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**INTERACTION OF OXYGEN AND GADOLINIUM
WITH Si(100)-2×1 SURFACE. FORMATION OF A SYSTEM
WITH 1-eV WORK FUNCTION**

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Changes in the electronic properties of the Si(100) surface, when a multilayer structure of oxidized Gd atoms is created on it, have been studied, by using the electron spectroscopy methods. It is shown that, after a number of adsorption cycles of Gd and oxygen atoms on the Si(100)-2×1 surface at room temperature and the annealing of the obtained structure at 600 °C, the work function decreases from 4.8 to less than 1 eV. The work function reduction at larger numbers of processing cycles is shown to be accompanied by the oxidation of Gd and Si atoms and a gradual decrease of the Si concentration in the near-surface region. The obtained results are explained by the formation of an O–Gd dipole layer on the surface.

Keywords: adsorption, gadolinium, oxygen, Si(100)-2×1 surface, oxidation, work function, dipole layer.

1. Introduction

Researches of the interaction between atoms of rare-earth elements and semiconductor surfaces is stimulated by the necessity to widen our knowledge concerning the properties of systems containing elements with unfilled f shells and to develop electronics and nanoelectronics, where those systems can be applied, e.g., Gd oxides and silicates with higher dielectric permittivities than that in SiO₂ [1], and so forth. Of interest is the Gd ability to form nanowires with unique properties on the Si surface [2–5], as well as the fact that gadolinium hexaboride has a low value of work function ($\varphi \approx 1.5$ eV) and is considered as a material for emission electronics [5, 6]. In connection with all that, the interaction of rare-earth elements, in particular, Gd, is studied rather intensively by various research groups [7–19]. In the cited works, it was found that Gd promotes the oxidation of the Si surface. The oxidation efficiency depends on the degree of Si surface coverage by Gd atoms, except for the cases where, owing to the annealing, the ordered Gd

silicides are formed on the surface [7, 8, 16]. The coadsorption of Gd and molecular oxygen on the Si surfaces, as well as the Gd adsorption on preliminarily oxidized Si surfaces, results in the formation of various Si-Gd-O interfaces [13, 18].

Recently, owing to the development of spintronics, it was found that Gd can be applied as a ferromagnet in new devices created on the basis of the spin-polarized tunneling [21], e.g., a spin metal-oxide-semiconductor field-effect transistor (spin MOSFET) with a semiconducting conductivity channel and the controllable ferromagnetic source and drain contacts [22]. The latter are suggested to be made from Gd using its ferromagnetism and its ability to form low-height Schottky barriers on silicon owing to the low value of work function of Gd itself [22, 23]. However, the studies of changes in the work function of such systems attracted little attention in the literature.

In particular, in work [14], it was reported that a Pr layer about 1 nm in thickness diminishes the surface work function of the system SiO₂/Si by 2.7 eV. In our previous works [24, 25], it was shown that the level-by-level adsorption of Gd and O atoms substan-

tially reduces the work function at the facet (113) of slightly doped silicon of the n -type to a value that is much lower than the characteristic work function of Gd. Layered Si–Gd–O systems on the Si(113) surface make it possible to obtain almost stable interfaces with a low work function $\varphi \approx 1$ eV. The work function of such a surface almost does not change under ultrahigh vacuum conditions and, after the specimen being held in the air environment, the indicated value can be restored by annealing the specimen at temperatures up to 600 °C. We proposed a hypothesis [25] that the reduction of φ originates from the creation of an O–Gd dipole layer near the surface, which is responsible for the formation of a surface with negative electron affinity. Unfortunately, the results of our researches did not give us a clear answer concerning the structure of the near-surface layer. The role of the structure, shape, and doping level of the used Si(113) surface was not clear. The role of the formation of ordered Gd silicide at the first stage was also obscured.

Taking into account that, in order to create the most efficient Si/O/Cs system with negative electron affinity, the Si(100) surface with degenerate p -doping was used [28], it is of interest to use the same Si surface in the Si/O/Gd system. Proceeding from all the aforesaid, this work was aimed at studying the level-by-level adsorption of atomic oxygen and gadolinium on the Si(100) surface with degenerate p -doping with the use of the valence-band photoelectron and Auger electron spectroscopy methods. The special attention was paid to variations of the work function on the surface of this system.

2. Experimental Techniques

As was done in our previous works [24–26], the researches were carried out in an ultrahigh vacuum chamber providing a base pressure of 2×10^{-10} Torr and equipped with special units for low-energy electron diffraction (LEED) measurements, photoelectron spectroscopy with a quantum energy in the interval $h\nu = 7.7\div 10.8$ eV, and Auger electron spectroscopy (AES). In a vacuum chamber, there were an ionic gun for cleaning the specimen surfaces using Ar ions, a system for the controllable deposition of Gd layers to a required coverage degree Θ_{Gd} , and a system for the specimen treatment with atomic oxygen to the given exposure $\xi = p(\text{pressure}) \times$

$t(\text{time})$. Molecular oxygen was introduced into the chamber. However, in the course of the exposition to the dose ξ , which was measured in Langmuir units ($1 \text{ L} = 10^{-6} \text{ Torr} \cdot \text{s}$), Si specimens were arranged at a distance of 1 cm from a tungsten wire heated up to about 2400 °C; O₂ molecules dissociated into atoms on the surface of this wire. Hence, in contrast to the previous researchers, we used the single-crystalline Si(100)- 2×1 facet as a substrate and atomic rather than molecular oxygen, thus excluding a complicated process of molecular oxygen dissociation on the silicon surface from consideration.

Valence-band photoelectron spectra (VB-PES) were measured with the help of a spherical analyzer of the bremsstrahlung type with a resolution of 0.1 eV. The energies of quanta for the photoelectron excitation were selected to equal 7.7 and 10.2 eV in order to make evaluation of the work function easier. The work function φ was calculated from the known relation $\varphi = h\nu - w$, where w is the energy of electrons excited from the Fermi level (E_{F}). It is defined as the energy distance from E_{F} to the low-energy edge of VB-PES. Since the system could be discontinuous [9, 10, 17, 23], the low-energy edge of VB-PES was extrapolated by a linear function [29], as is illustrated in Fig. 2. Si(100) specimens were cut out from boron-doped wafers ($\rho \approx 0.02 \Omega \cdot \text{cm}$). After a preliminary chemical treatment, the specimens were cleaned in the vacuum chamber with the help of several cycles of their bombardment with argon ions and annealed at a temperature of 600 °C. After cleaning the specimen under ultrahigh vacuum conditions, distinct spots corresponding to a 2×1 superstructure were registered on the Si(100) surface using the LEED technique, whereas Auger spectra did not register its appreciable contamination with oxygen and carbon. The cleaned surface was deposited with Gd to a coverage degree of about 6 ML. After the system was annealed at 600 °C for 5 min, Gd silicide was formed at the surface with a distinct 2×1 pattern of LEED spots and the corresponding AES and VB-PES curves (curves b in Figs. 1 and 2). The initial level of Gd coverage and the annealing temperature were selected to obtain the initial system in a steady state, which followed from our previous researches [24, 25]. The initial specimen was repeatedly exposed to atomic oxygen to doses sufficient for the Si oxidation to saturate [7, 9]. Then it was deposited with a Gd layer and annealed. The annealing

temperature was selected to equal $T \approx 600$ °C. At this temperature, no substantial changes in the interface composition were observed, which was monitored with the help of AES [15]. Moreover, in accordance with the results of work [5], Gd does not evaporate from the Si surface even at 1200 °C, but only diffuses into the bulk and, after the cooling, segregates on the surface. After the first exposition in oxygen (250 LO), the surface of Gd silicide became disordered, and no LEED spots were registered on the surface of the examined systems after the further annealings and Gd depositions.

3. Research Results

In Fig. 1 and 2, typical AES and VB-PES are shown for one of the cycles (Si(100) + 6 ML Gd + 600 °C) of treatment of the specimen. They illustrate changes that occur on the specimen surface (curves *b* to *k*) after every exposition in atomic oxygen, deposition of additional Ga layers, and annealing. For comparison, curves *a* in the same figures demonstrate the corresponding spectra for the initial Si(100)-2 × 1 surface. By comparing curves *a* and *b* in Fig. 1, we may draw conclusion that, after depositing 6 ML of Gd on Si and annealing at 600 °C, the Auger signal Si(LVV) decreases, and the characteristic Auger lines inherent to Gd(NON) and Gd(NVV) transitions appear in the energy interval of 100–160 eV. The attention is attracted by the form of Gd(NVV) transitions. From curves *b* in Fig. 1 and in inset B in Fig. 1, one can see that this is a comb of three peaks (at 125, 130, and 140 eV). Their shape corresponds to that quoted for Gd in the handbooks of Auger spectra [16, 27]. The VB-PES of formed silicide (curves *b* in Fig. 2) is characterized by a considerable density of states near E_F . The silicide work function was determined to equal about 4 eV. The appearance of a low-energy shoulder in the VB-PES (curves *b*) can be associated with the formation of a discontinuous structure on the surface.

After the first exposition of the specimen in atomic oxygen (250 LO), the Auger and photoelectron spectra of the system strongly changed (curves *c* in Figs. 1 and 2, and in inset A in Fig. 1). In the Auger spectrum, the intensity of the Auger line of clean Si (at 92 eV) decreased, but the Auger line of oxidized Si (at 68 eV) emerged. The shape of the Gd(NVV) line changed in such a way that the peak at about 125 eV

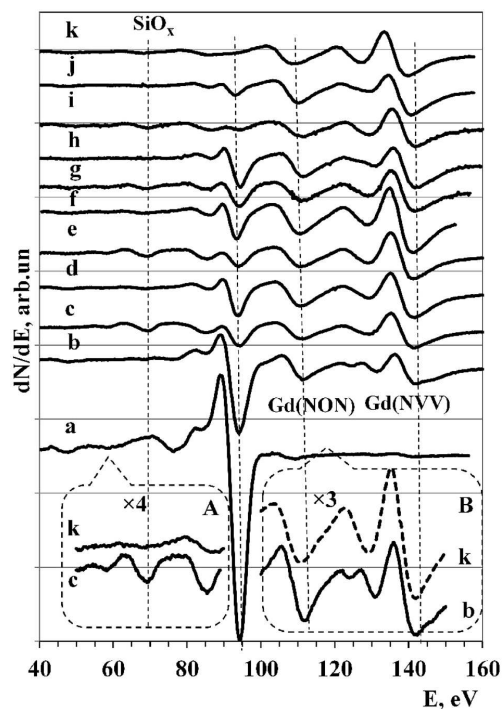


Fig. 1. Evolution of Auger spectra for the system Si(100)/Gd/O: *a*: Si(100); *b*: *a*+6 ML Gd + 600 °C; *c*: *b* + ($\xi = 250$ LO); *d*: *c* + 2 ML Gd ($\Sigma\Theta_{\text{Gd}} = 8$ ML); *e*: *d* + 100 LO ($\Sigma\xi = 350$ LO); *f*: *e* + 2.5 ML Gd ($\Sigma\Theta_{\text{Gd}} = 10.5$ ML); *g*: *f* + 600 °C; *h*: *g* + 2 ML Gd ($\Sigma\Theta_{\text{Gd}} = 12.5$ ML); *i*: *h* + ($\Sigma\xi = 450$ LO) + 600 °C; *j*: after $\Sigma\xi = 550$ LO and $\Sigma\Theta_{\text{Gd}} = 18.5$ ML; *k*: after $\Sigma\xi = 650$ LO, $\Sigma\Theta_{\text{Gd}} = 19.5$ ML+600 °C. Scaled-up Auger spectra *c* and *k* in the interval 50–90 eV are shown in inset A. Scaled-up Auger spectra *b* and *k* in the interval 100–150 eV are shown in inset B

disappeared, which can be associated with the oxidation of Gd [25].

In the following cycles of deposition of additional Gd layers (curves *d* and *f*), when the specimen was exposed to oxygen before the deposition and, afterward, annealed, the magnitude of Auger lines Gd(NON) and Gd(NVV) changed weakly, but the shape of Gd(NVV) lines corresponded to the shape of lines for oxidized Gd atoms. In addition, the intensity of Auger lines Si(LVV) considerably increased, and the intensity of the Auger line of oxidized silicon (at 68 eV) decreased. At the same time, after the deposition of those additional Gd layers (curves *d* and *f*), states at $E_F - 1.8$ eV appeared in the VB-PES, and no additional states appeared up to E_F . The work function in the system with $\Theta_{\text{Gd}} = 10.5$ ML (curves *f*

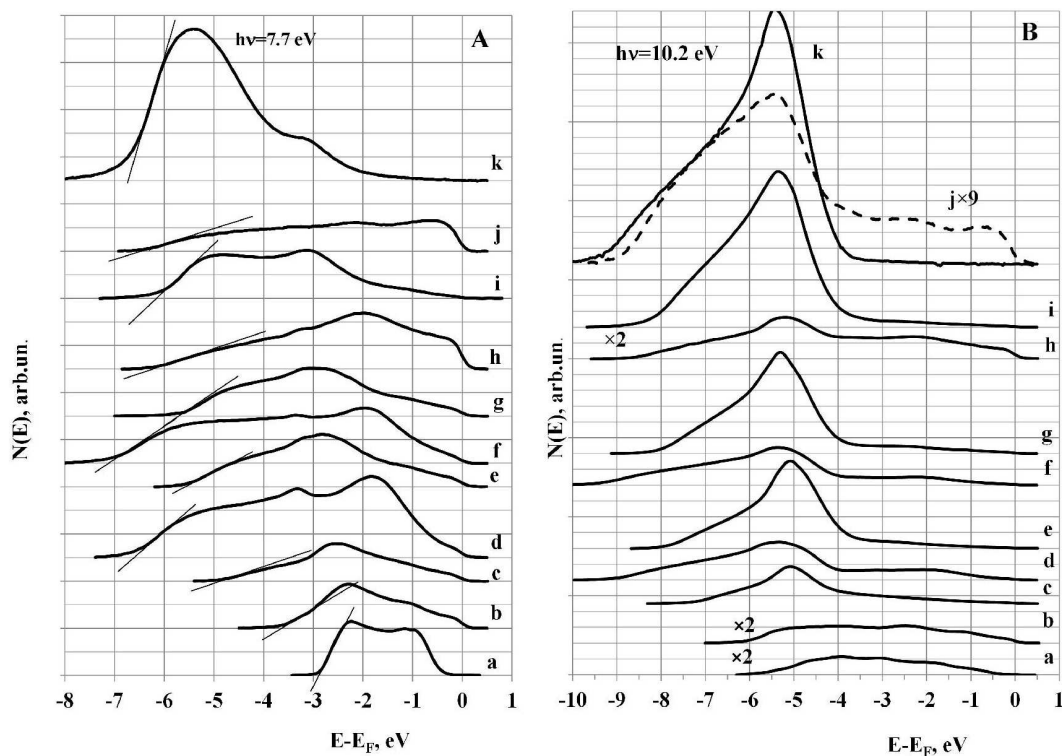


Fig. 2. (VB-PES of Si(100)/Gd/O interface: $h\nu = 1.7$ (A) and 10.2 eV (B). Curve notation is the same as in Fig. 1

in Figs. 2) amounted to about 0.7 eV. However, such a low value for the work function was not stable and, at heating, increased almost to 2 eV (curves *g* in Figs. 2). All described facts allow us to consider that, at those stages, additionally introduced Gd atoms diffuse into the bulk, break the oxygen bonds with silicon, and become oxidized, which does not contradict the literature data [7, 9].

Starting from the total thickness of a deposited layer $\Theta_{\text{Gd}} = 12.5$ ML (curves *h* in Figs. 1 and 2), the sputtering of every new portion of Gd atoms gave rise to the appearance of a peculiarity in the Auger spectra at about 125 eV, and rather a considerable density of states appeared in the photoelectron spectra near E_F (curves *h* and *j* in Figs. 1 and 2). Those peculiarities disappeared after the specimens had been exposed to atomic oxygen. Hence, every exposition cycle of the examined specimens in atomic oxygen was accompanied by a reduction in the intensity of the Auger line LLV of clean Si at 92 eV, the appearance of the line of oxidized Si in the energy interval about 68 eV, the disappearance of the peak near

125 eV in the Auger spectra at the density of metallic states near E_F in VB-PES, which, as was supposed in our previous work [25], is caused by the Gd oxidation. In other words, we have all grounds to suppose that, after the exposition in atomic oxygen, the partial oxidation of Gd and Si atoms near the surface took place, as well as the embedding of oxygen atoms under the surface Gd atoms. As a result of such embedding, a complicated dipole layer (O–Gd or Si–O–Gd) emerged near the surface, which reduced the surface work function in the system. Under the influence of oxygen, the VB-PES also underwent substantial modifications: the density of states in the energy interval around $E_F - 5$ eV grew, and the integrated photosensitivity of the interface (the area under the VB-PES curves) increased.

After several cycles of the treatment ($z > 10$)—every of them consisted of the deposition of additional Gd layers, exposition to atomic oxygen, and annealing at 600 °C—when the total exposure dose reached $\xi \approx 650$ LO and at the additional annealing, a rather stable system was formed, the AES and VB-PES of

which are exhibited by curves k in Figs. 1 and 2. Its Auger spectrum is characterized by the lines of oxidized Gd (cf. curves b and k in inset B in Fig. 1), the almost zero contribution from the Auger line of clean Si (92 eV), and an insignificant contribution from oxidized Si (curves k in Fig. 1 and in inset A in Fig. 1), which is determined by a lower intensity of the Auger line with an energy of 68 eV. The VB-PES of this stable system reveals an intense spectral peculiarity with a maximum in the energy interval ($E_F - 5.5$ eV, $E_F - 5.3$ eV), which, in accordance with the literature data [9, 10, 19], can be attributed to the states of oxygen $2p$ electrons, and a much less intense spectral peculiarity at $E_F - 3.2$ eV.

Our data are qualitatively similar to those available in the literature [10], which were obtained for the Si(111)-Gd interface oxidized in molecular oxygen. However, we attract attention to the following facts: (1) our photoelectron spectrum is rather wide, i.e. the work function of this system becomes almost one-fifth as high as the work function of silicon; it is evaluated to be approximately 1 eV; (2) the states near E_F disappear; and (3) the photosensitivity of the obtained system grows owing to an increase in the number of photoelectrons with low kinetic energies, which means a nonmetallic character of the system. Together with the authors of work [10], we consider that the complete absence of states near the Fermi level testifies that the near-surface region becomes almost insulating, with an energy gap width no less than 4 eV.

4. Discussion of Results

In order to illustrate the evolution of the examined specimens, the dependences of the intensities of Auger lines Si(LVV) ($I_{\text{Si(LVV)}}$), Gd(NON) ($I_{\text{Gd(NON)}}$), and O(KLL) ($I_{\text{O(KLL)}}$), as well as the work function φ , on the operation cycle number z are depicted in Fig. 3. The signal $I_{\text{Si(LVV)}}$ was normalized by the magnitude of the corresponding Auger signal from the clean Si surface ($z = 0$ in Fig. 3), the signal $I_{\text{Gd(NON)}}$ by the magnitude of the corresponding Auger signal from the first of six Gd monolayers on the Si(100) surface ($z = 1$ in Fig. 3), and the signal $I_{\text{O(KLL)}}$ by the magnitude of the corresponding Auger signal after the first exposition in oxygen to 250 LO ($z = 3$ in Fig. 3). The work function φ was normalized to the work function $\varphi = 4.8$ eV of

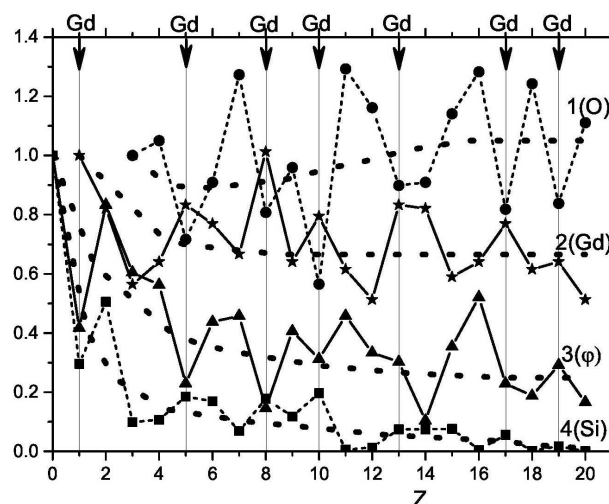


Fig. 3. Dependences of the normalized intensities of Auger lines O KLL (1), Gd NON (2), Si LVV (4), and the work function φ (3) on the number of cycles of treatment: $z = 0$: atomically clean Si(100) surface; 1: Si (100) + 6 ML Gd; 2: annealing at 600 °C; 3: exposition to $\xi = 250$ LO; 4: annealing at 600 °C; 5: 4 + 2 ML Gd; 6: annealing at 600 °C; 7: $\xi = 100$ LO; 8: 7 + 2.5 ML Gd; 9: annealing at 600 °C; 10: 9 + 2 ML Gd; 11: $\xi = 100$ LO; 12: annealing at 600 °C; 13: 12 + 3 ML Gd; 14: holding in ultrahigh vacuum for 6 days; 15: annealing at 600 °C; 16: $\xi = 100$ LO; 17: 16 + 3 ML Gd; 18: $\xi = 100$ LO and annealing at 600 °C; 19: 18 + 1 ML Gd; 20: annealing at 600 °C

the atomically clean Si(100)-2×1 surface ($z = 0$ in Fig. 3). The Si(100) + 6 ML Gd interface was selected as the initial specimen ($z = 1$). Later, it was cyclically treated by the annealing at about 600 °C, exposition to atomic oxygen, and deposition of additional Gd layers. Within every cycle, the sequence of the annealing and the exposition to oxygen could be changed. Nevertheless, one can see (Fig. 3, curve 4) that, as the number of cycles, z , grows, a general tendency is observed that the intensity of Auger line Si(LVV) decreases within the interval of Auger electron escape depth (about 1 nm), which is natural because the amount of gadolinium and oxygen increases at the interface surface, in contrast to the clean Si surface. At the same time, the number of Si atoms a little fluctuates against the background of its general decrease, because there are the processes of silicon and gadolinium oxidation in the specimen, as well as their diffusion-driven mixing and segregation, which does not contradict the literature data [7]. The intensity ratio $I_{\text{Gd(NON)}/I_{\text{Gd(NON)}}$ (Gd 6 ML)

(Fig. 3, curve 2) fluctuates with a considerable spread around the average value of about 0.65, and the $I_{O(KLL)}/I_{O(KLL)}(250\text{ LO})$ one (Fig. 3, curve 1) fluctuates around 1. This means that, at $z < 10$, the interface becomes enriched with oxygen. The final interface consists of gadolinium silicides, as well as oxides with Gd and Si admixtures. Our results agree with the literature data, but the dependence of the interface work function on the operation cycle number z (Fig. 3, curve 3) is new. The work function decreases, on the average with a certain spread, by almost a factor of 4.8 to a value of about 1 eV. One can also see that, at the given z , it becomes less than the average value, when the $I_{O(KLL)}/I_{O(KLL)}(250\text{ LO})$ -values are minimum and the $I_{Gd(NON)}/I_{Gd(NON)}(\text{Gd } 6\text{ ML})$ -values are maximum (in Fig. 3, the arrows mark the numbers of cycles z , when additional Gd layers were deposited). When additional Gd layers are deposited, the intensity of the Auger line Si(LVV) increases with respect to the average value (curve 4 in Fig. 3). This circumstance can be explained by the fact that some fraction of the additional amount of gadolinium becomes oxidized and takes away some part of oxygen from silicon oxide. After the exposition in oxygen, the character of the interface changes from metallic, which takes place every time, when the Gd amount exceeds its average value, to semiconducting or even insulating, when the amount of oxygen increases. At $z > 15$, an almost stable interface consisting of oxides and Gd silicides is formed. This interface has a complicated dipole layer composed of Gd–O on its surface, which is similar to the model proposed for the explanation of a considerable reduction in the work function of gallium arsenide with the use of Cs₂O dipoles [26], and is responsible for a considerable reduction of the work function to a value less than 1 eV. The work function of the formed surface held in ultrahigh vacuum for a long time slightly increases (by about 0.5 eV), but comes back to a value less than 1 eV after the annealing at 300–600 °C. Holding the specimen in air increases the work function to about 4 eV, but annealing under the ultrahigh vacuum conditions diminishes the work function to values less than 1 eV again. It is very probable that the researched system is discontinuous, which complicates its analysis [7, 9, 15]. When selecting the Si(100) surface with degenerate p -doping for researches, we expected that the influence of Gd and O should result in the for-

mation of a surface with the work function much lower than the value obtained by us in work [25], where the facet (113) of weakly doped n -silicon was used as a substrate. However, after the treatment with oxygen and gadolinium, the surface work function turned out almost identical in both cases. This fact can be explained by the presence of oxygen, which, being mainly an acceptor, is responsible for the formation of a semiconductor structure of the p -type with E_F fixed near the valence band top. Hence, there emerges a structure similar to those formed on p -semiconductor surfaces at the adsorption of Cs and O and characterized by a negative electron affinity.

5. Conclusions

The researches of cycles with the interaction between gadolinium and atomic oxygen at their level-by-level adsorption on the Si(100) surface and the annealing, which were carried out with the help of LEED, AES, and VB-PES methods, made it possible to trace the evolution in the formation of complicated interface, possibly discontinuous, with a considerably reduced work function (<1 eV). The interface is composed of Gd, its silicides and oxides, and Si. It turned out that the character of the interface with various relative amounts of Gd changes from metallic to semiconducting. Every time when, at the Gd adsorption, the near-surface region becomes more metallic, the relative amounts of Gd and Si in it increase, whereas the relative amount of oxygen decreases. If this system is oxidized, a complicated dipole layer consisting of O–Gd is formed in the near-surface region, which efficiently reduces the surface work function. As the number of cycles including the level-by-level adsorption and the annealing increases, the relative amount of Si gradually decreases, and the interface becomes more semiconducting, more stable, and more photosensitive. The surface work function gradually decreases down to about 1 eV, which is typical of surfaces with a negative electron affinity.

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ВЗАЄМОДІЯ КИСНЮ
ТА ГАДОЛІНІЮ З Si(100)-2×1. УТВОРЕННЯ
СИСТЕМИ З РОБОТОЮ ВИХОДУ 1 eV

Резюме

Методами електронної спектроскопії досліджено зміну електронних властивостей поверхні Si(100) при створенні на ній багатопарової структури окислених атомів Gd. Показано, що внаслідок ряду циклів адсорбції атомів Gd та атомарного кисню при кімнатній температурі на поверхню Si(100)-2×1 та відпалу отриманої структури при ≈600 °C робота виходу поверхні зменшується від 4,8 eV до значень, менших від 1 eV. Встановлено, що зменшення роботи виходу зі збільшенням циклів обробки супроводжується окисленням атомів Gd та Si і поступовим зменшенням концентрації Si в приповерхневій області. Отримані результати пояснюються утворенням на поверхні дипольного шару O—Gd.