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INVESTIGATION OF VARIOUS MECHANISMS OF RADIATIVE RECOMBINATION OF SILICON NITRIDE AS AN EFFECTIVE WAY TO BROADENING THE PHOTOLUMINESCENCE SPECTRUM

Abstract

The development of devices combining optical and electrical functions based on silicon-containing materials is one of the challenges in microelectronics. By plasma enhanced chemical vapor deposition synthesis and subsequent annealing, silicon nitride samples with both excess silicon and excess nitrogen were formed. The high concentration of Si-H and N-H bonds was determined by Raman spectroscopy in samples before annealing. By the transmission electron microscopy, it was determined that in addition to silicon nitride, silicon clusters were formed in the sample's matrix. The photoluminescence spectra changed significantly for both types of samples during annealing in different gas atmospheres. Heat treatment of samples at 1100 °C after synthesis led to the disappearance of the PL spectrum, and after annealing at 800 °C, photoluminescence increases. It is noted that the highest intensity of photoluminescence was detected after annealing in the air atmosphere and the lowest in the nitrogen. The participation of N centers in recombination processes was confirmed by the method of electron paramagnetic resonance. The different mechanisms of particle interaction leading to photoluminescence and charge storage are considered. Thus, the conditions for the synthesis and annealing of silicon nitride layers are selected to obtain controlled luminescent properties in various spectral ranges.

Key words: silicon nitride, silicon oxide, luminescence, nanoclusters, stoichiometric order.

Introduction

The prospects for the development of silicon photonics are based primarily on the implementation of an optical data transmission system in an integrated circuit [1–2]. Currently, almost all components

of silicon optoelectronics have been created. The complexity of obtaining a working LED based on Si is due to its non-direct-band energy structure. To solve this issue, the formation of Si nanocrystals in various matrices is also applicable. One of the standard methods for obtaining Si nanocrystals is based on the heat treatment of silicon oxide enriched with silicon.

Due to the smaller band gap (~ 5.3 eV), silicon nitride is a more suitable basis for the development of electroluminescent devices compared to wide-band silicon oxide SiO_2 (8.2 eV) [3]. Silicon nitride has a lower barrier at the cluster/matrix interface and therefore more efficient injection of charge carriers into silicon nanocrystals [4].

However, the silicon nitride matrix also luminesces, and the nature of the radiation is widely discussed. The emissivity of silicon nitride is associated with electronic transitions between the energy levels of defects [5–9]. The main centers of radiation recombination in this case are the K- and N-centers [10]. The K-center level is located close to the middle of the silicon nitride band gap, and the nature of this center is amphoteric (both electrons and holes can be captured). Consequently, electronic transitions are possible both between the valence band and the K-center, and between the K-center and the conduction band.

The levels of the N-centers are located closer to the edges of the bands: the N_2^0 level is ~ 0.5 eV higher than the top of valence band, the N_4^+ level is closer to the bottom of the conduction band. According to some data, their levels may be even below the edge of the valence band [11, 12]. In this case, the centers are inactive in radiative recombination. However, the model mentioned above demonstrates the possibility of a negative transition between the N_2^0 level and the conduction band (3.1 eV) and a transition between the N_4^+ level and the valence band (3.0 eV).

Comparing various mechanisms of energy transfer involving the main centers of silicon nitride - K and N, in particular when emitting light quanta, the efficiency of K centers is obviously higher. By increasing the concentration of K-centers, we increase the capacity of a memory cell. Another mechanism is associated with the localization of a hole in the field of an electron trapped at the K center. Such a structure can retain properties under certain conditions.

In the plasma enhanced chemical vapor deposition (PECVD) process, it is possible to influence the concentration of Si-H and N-H bonds, which, when broken, form structures with uncompensated charge. By changing the annealing modes (atmosphere, temperature, etc.), both silicon clusters and various luminescent centers can be obtained.

In a sample with an excess of silicon, a two-phase structure of silicon nitride and silicon clusters can be formed as a result of synthesis and subsequent annealing. There will probably be several photoluminescence (PL) sources in this system.

The novelty of this study lies in finding conditions for the synthesis and annealing of silicon nitride at which luminescence increases associated with radiative centers, silicon clusters and states in the band tails.

The motivation of the research was the possibility of broadening the PL spectrum of silicon nitride in the implementation of various light-emitting mechanisms.

An important feature of this work is the obtaining of silicon nanoclusters both in silicon nitride samples with an excess of silicon and with an excess of nitrogen.

Thus, considering most materials at the level of interaction of particles with uncompensated charge is a cornerstone in understanding the mechanisms of photoluminescence and many other issues.

Materials and methodology

The Si-rich and N-rich silicon nitride films were deposited on the n-type (100)-oriented Si substrates by plasma-enhanced chemical vapor deposition at 350 °C using a gaseous mixture of silane (SiH_4) and nitrogen (N_2) as precursors. The ratio of reagent gases SiH_4/N_2 was chosen 1/3 for the Si-rich and 1/6 for N-rich silicon nitride films. The pressure in the chamber was maintained at 70 Pa.

The activation energy of the silicon nitride deposition process is approximately 1.8 eV. The deposition rate of silicon nitride usually depends on the concentration of silane and ammonia in the reaction mixture as well as the total pressure in the system [13–17].

Plasma enhanced chemical deposition makes it possible to form films at a very low substrate temperature. This is achieved due to the reaction between gases in a glow discharge. Although the temperature of the electrons in the discharge can reach 105 °C, the sample temperature remains within 100–400 °C. This is undoubtedly important in the manufacture of semiconductor devices, since it does not cause the formation of excessive defects.

Films obtained by PECVD contain a large amount of hydrogen (10–35 %_{at}), the concentration of which depends on the parameters of the deposition process [18]. Hydrogen in the films forms a bond with silicon in the form of Si–H and with nitrogen in the form of N–H [19]. Also, plasma-chemical silicon nitride contains 0.5–2 %_{at} oxygen as an impurity.

In the presented work, the Raman spectra were recorded using a "Nanofinder HighEnd (LOTIS TII)" micro-Raman spectrometer. The studies were carried out at room temperature. The radiation source was a laser with $\lambda = 473$ nm. The laser radiation was focused into a spot with a diameter of ~ 1 μ m. Diffraction gratings of 600 strokes/mm (spectral resolution 0.01 nm) was used for registration.

Structural-phase transformations in silicon nitride films were studied by TEM on a Hitachi H-800 microscope with an accelerating voltage of 200 keV in the "cross-section" geometry. The resolution of the microscope is 0.2 nm, magnification is 100–600000.

The PL spectra of silicon nitride films in the visible region were recorded using a photomultiplier tube sensitive in the spectral region 200–900 nm (Hamamatsu Company, Japan). A UV He–Cd laser with a wavelength of 325 nm was used to excite photoluminescence in the spectral range of 350–800 nm and an Nd laser with a wavelength of 532 nm was used to excite photoluminescence in the range of 600–900 nm. Transitions between the levels of K and N centers and the level of the conduction band were excited.

EPR spectra were recorded at room temperature using a JES -FA (Jeol) spectrometer and a reference value of Mn(2+)/MgO. The spectra were accumulated four times in order to raise the S/N ratio.

Results and discussion

In this paper, the objects of research are two series of samples of non-stoichiometric silicon nitride with a lower Si concentration in SiN_x – III-10 and with a higher Si concentration in SiN_x – III-13. A thin layer of silicon nitride was deposited on two silicon wafers by PECVD at a substrate temperature of 350 °C, the characteristics of which are presented in Table 1.

Table 1 – Characteristics of silicon nitride layers deposited by PECVD on silicon wafers

Characteristics of the silicon nitride layer	Plate number	
	III-10	III-13
SiH ₄ /N ₂ ratio	1/6	1/3
Thickness, μ m	0,34–0,36	0,3–0,32
Stoichiometric parameter	1,5–1,41	1,13

For heat treatment, furnace annealing was used for 60 minutes in three atmospheres – argon (Ar), nitrogen (N₂) and in air using two temperatures of 800 °C and 1100 °C for both series.

The low synthesis temperature leads to the formation of bonds with hydrogen. Further treatment of the obtained samples at various parameters allows the formation of various structures, including light-emitting ones.

An important condition for the formation of effective light-emitting centers is the presence of hydrogen bonds after synthesis and their subsequent annealing during heat treatment. The formation of Si–H and N–H bonds in the PECVD process was confirmed by Raman spectroscopy. Figure 1 shows the spectrum for a sample with excess nitrogen. As can be seen from the figure, the bands with maxima at 2230 cm^{-1} and 3330 cm^{-1} correspond to fluctuations of Si–H and N–H bonds in the initial sample [20–22]. After heat treatment, these bands are not registered in the spectrum. The disappearance of bands from the spectrum is associated both with the dissociation of these bonds and with an increase in the luminescent background. For a sample with an excess of silicon, the Raman signal from Si–H and N–H bonds was suppressed by a strong luminescence background.

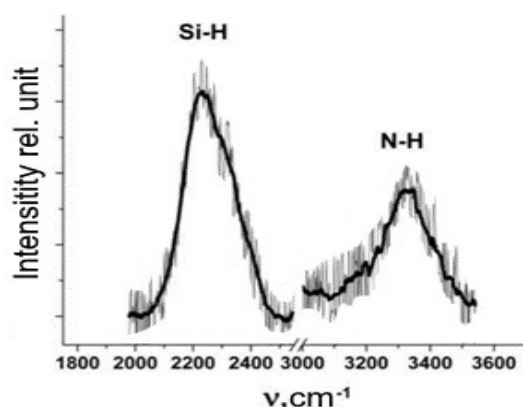


Figure 1 – Raman spectrum for a sample with excess nitrogen

After annealing of the samples, hydrogen-containing bonds are broken and stable light-emitting centers are formed. Their presence was determined by the characteristic peaks in the photoluminescence spectra.

The interaction of particles with uncompensated charge, leads to the formation of clusters. Such structures have a higher intensity of PL than individual particles. To determine the formation of clusters, TEM images of all types of samples were obtained.

Figure 2 shows TEM images of samples with excess silicon and nitrogen, annealed at 800 and 1100 °C. Image analysis indicates the presence of some inhomogeneities in annealed silicon nitride films. As can be seen, nitride films annealed at 800 °C, both with an excess of silicon or nitrogen, contain small contrast inhomogeneities. After annealing at a temperature of 1100 °C, the uniformity of the structure of the nitride film increases significantly.

In [23] states that silicon nitride films with a high content of silicon, both initial and annealed at $T < 650$ °C, consist of a mixture of two phases: amorphous silicon nitride with an excess of silicon and with an excess of nitrogen. Only after annealing at $T > 800$ °C, the formation of silicon clusters in a matrix of amorphous silicon nitride was detected. Most possible, the inhomogeneities of the structure are associated with the separation of phases in the film of amorphous silicon nitride into regions with an excess of silicon $\text{SiN}_{x=1.3}$. Annealing at a temperature of less than 800 °C leads primarily to the break of weaker Si–H and N–H bonds. It is obvious that the concentration of Si–H bonds is higher in areas enriched with silicon. Consequently, annealing at a relatively low temperature (less than 800 °C) will lead to hydrogen effusion and a decrease in density in these areas. Thus, the lower contrast of the images is probably due to the areas enriched with silicon. Heat treatment at higher temperatures (800–1100 °C) leads to annealing of defects such as broken bonds through their interaction. This mechanism leads to the synthesis of silicon clusters. This, in turn, should lead to smoothing of the heterogeneous structure, which is observed in the TEM images.

The variety of different structures, clusters and particles with uncompensated charge leads to different manifestations of their activity when excited by light.

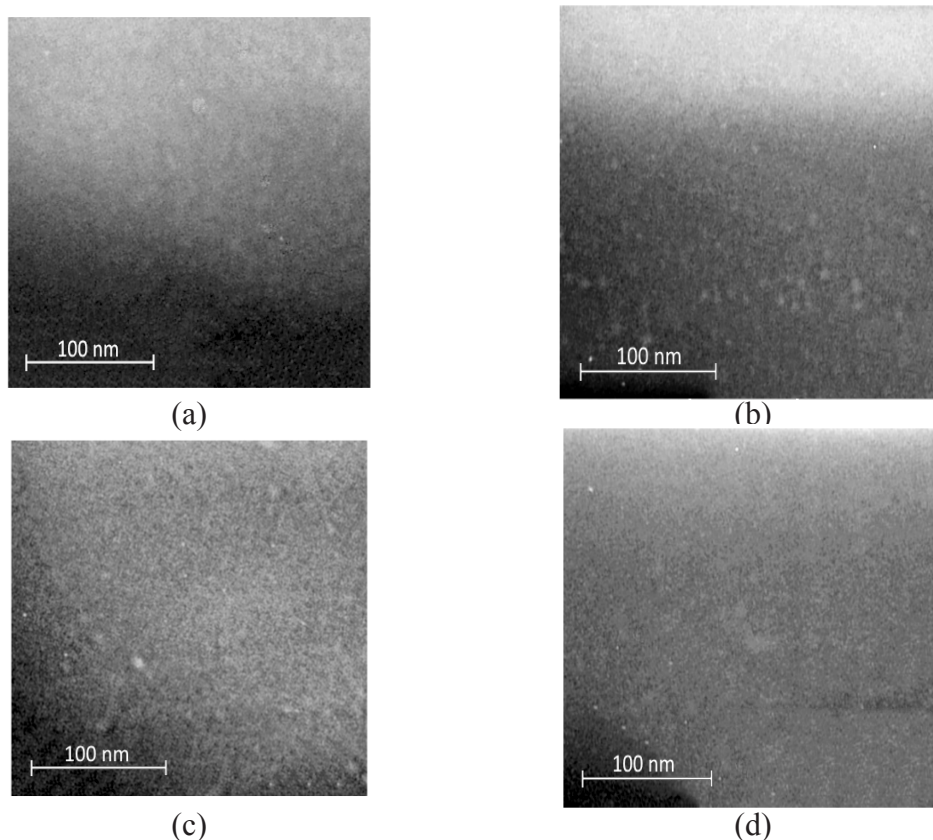


Figure 2 – TEM micrographs of the «cross-section» geometry of samples annealed in Ar for an hour.
Figures a, b – $T_{\text{anneal}} = 800\text{ }^{\circ}\text{C}$; c, d – $T_{\text{anneal}} = 1100\text{ }^{\circ}\text{C}$; a, c – a sample with an excess of silicon;
b, d – a sample of a series with an excess of nitrogen.

Figure 3 shows the photoluminescence spectra of the two initial samples.

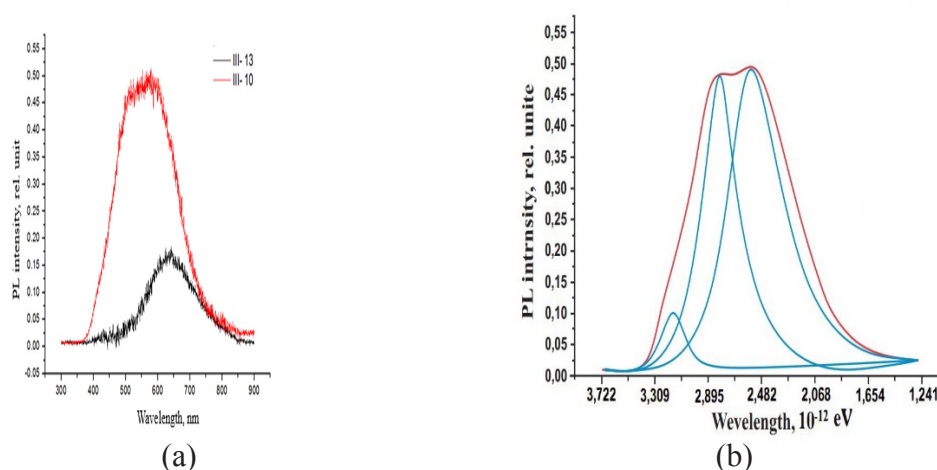


Figure 3 – a – Photoluminescence spectra of initial samples,
b – The spectrum for the III-10 series sample decomposed into Gaussians

For the series – III-10 non-stoichiometric SiN_x , a wide peak is observed, which is at around 0.52 relative units, in the range of 580 nm, this is approximately 2.14 eV. The width at half-height is 460-670 nm (2.7–1.85 eV). This spectrum also has a second weak peak – at a wavelength of 410 nm

(3.02 eV), with an intensity of 0.255 rel. units. The peak of the second spectrum of the SiN_x series – III-13 has a lower intensity – 0.17 rel. units and is located at 640 nm (1.94 eV). It is shifted relatively to the first peak in the long-wavelength region. This spectrum has another weak peak – 430 nm (2.88 eV) with a intensity of 0.06 rel. The unit shift of the spectral line to the long-wavelength region indicates a higher Si content in SiN_x series III-13 compared to series III-10.

Figure 4 shows the change in the position of the spectral bands of the III-10 series as a result of annealing in argon compared to the initial one. During heat treatment at 800 °C, the peak of the spectrum becomes sharper, shifts to the short-wave region to a value of 510 nm (2.43 eV) with an intensity of 0.76 rel. units. The width at half-height is 440-610 nm (2.82–2.03). At a temperature of 1100 °C, photoluminescence is significantly extinguished and already amounts to 0.18 rel. units at 640 nm (1.94 eV). The width at half-height is 470–700 (2.64–1.77).

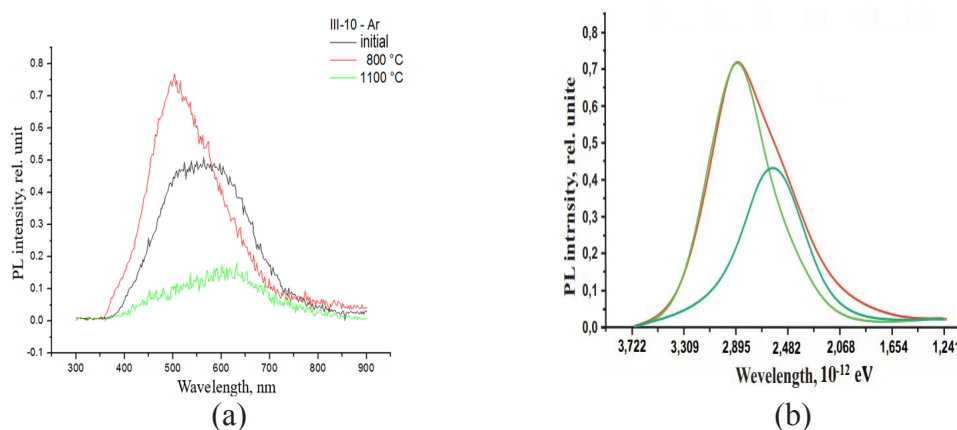


Figure 4 – a – Photoluminescence spectra of the initial sample and after annealing in argon for series III-10;
b – The spectrum of the initial sample of series III-10 decomposed into Gaussians

The energy interval between the K center and the conducting band (CB) is less than that between the N center and CB. Therefore, K centers are more actively involved in radiative recombination. Based on this, methods with increased sensitivity, such as EPR spectroscopy, are needed to determine the properties of N centers.

For the sample of series III-13, after annealing in a nitrogen atmosphere at 800°C (Fig. 5), two lines caused by unpaired electrons were observed in the EPR spectrum. The choice of this sample is due to the highest intensity of PL. Characteristics of the first EPR line: g – factor = 2.00173, width $\Delta H = 8$ G. This line could not be identified.

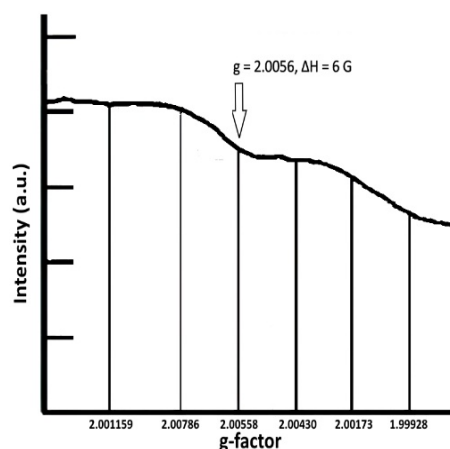


Figure 5 – EPR spectrum of the III-13 series sample after annealing at 800 °C in a nitrogen atmosphere

Characteristics of the second line: g – factor = 2.0056, width $\Delta H = 6$ G. In [24], a line in the EPR spectrum of amorphous silicon nitride with similar characteristics (g – factor = 2.0057, width $\Delta H = 8.5$ G) was attributed to a 2-coordinated nitrogen atom (N-center). Therefore, we can talk about the presence of N-centers in the samples, and the increased luminescence of this sample can be explained by the recombination of carriers in the N-centers.

Conclusion

During the PECVD processes, various light-emitting structures of silicon nitride were formed by changing the SiH_4/N_2 ratio. The Raman spectroscopy method revealed the intensive formation of Si-H and N-H bonds, which, upon further annealing, are broken during the formation of light-emitting centers. The synthesis of silicon clusters after annealing of samples with an excess of silicon and nitrogen was determined from TEM images. The mechanisms of cluster formation vary significantly depending on the features of the interaction of particles with uncompensated charge.

The influence of the atmosphere and the temperature of annealing on the efficiency of photoluminescence of the formed layers was determined. During annealing at 1100 °C, the intensity of all samples is significantly reduced, and for initial samples with a higher silicon content, it is completely absent. During annealing at 800 °C, photoluminescence increases.

For each annealing mode, the mechanism of formation of radiative particles is determined, as well as their effect on the intensity of PL. Radiative recombination is associated with the presence of K and N centers, silicon clusters, and edge luminescence.

The different silicon content in the initial nitride samples determines the spectral position of the photoluminescence band. The higher silicon content corresponds to the shift of the maximum of the PL band from 580 nm to 640 nm. In our opinion, the radiative recombination of electrons and holes in this case occurs between states (energy levels) in the tails of the energy bands.

The detected PL bands in the short-wave region can be associated with the formation of Si nanoclusters. An increase in the heat treatment temperature leads to an increase in the size of nanoclusters, which, in turn, leads to a red shift of the photoluminescence bands.

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ФОТОЛЮМИНЕСЦЕНЦИЯ СПЕКТРІН КЕҢЕЙТУДІҢ ТИІМДІ ӘДІСІ РЕТІНДЕ КРЕМНИЙ НИТРИДІНІҢ РАДИАЦИЯЛЫҚ РЕКОМБИНАЦИЯСЫНЫҢ ӘРТҮРЛІ МЕХАНИЗМДЕРІН ЗЕРТТЕУ

Андатпа

Құрамында кремний бар материалдар негізінде оптикалық және электрлік функцияларды біріктіретін құрылғыларды әзірлеу — микроэлектрониканың негізгі міндеттерінің бірі. Химиялық тұндыру күшейтілген плазмалық синтез арқылы газ фазасынан және одан кейінгі күйдіруден артық кремниймен де, артық азотпен де байытылған кремний нитридінің үлгілері алынды. Si-H және N-H байланыстарының жоғары концентрациясы күйдіру алдындағы үлгілерде Раман спектроскопиясы әдісімен анықталды. Трансмиссиялық электронды микроскопия арқылы кремний нитридінен басқа, үлгі матрицасында кремний кластерлерінің пайда болатыны анықталды. Фотолюминесценция спектрлері әртүрлі газ атмосфераларында күйдіру кезінде үлгілердің екі түрі үшін де айтарлықтай өзгерістер көрсетті. Синтезден кейін 1100 °C температурада үлгілерді термиялық өңдеу PL спектрінің жоғалуына әкелді, ал 800 °C температурада күйдіргеннен кейін фотолюминесценцияның қарқындылығы жоғарылады. Фотолюминесценцияның ең жоғары қарқындылығы ауа атмосферасында күйдірілгеннен кейін байқалды, ал азот атмосферасында ең төменгі қарқындылық анықталды. N-орталықтардың рекомбинациялық процестерге қатысуы электронды парамагниттік резонанс әдісімен расталды. Фотолюминесценцияға және зарядтың жиналуына әкелетін бөлшектердің өзара әрекеттесуінің әртүрлі механизмдері қарастырылды. Осылайша, әртүрлі спектрлік диапазондарда басқарылатын люминесценттік қасиеттерді алу үшін кремний нитриді қабаттарын синтездеу және күйдіру шарттары таңдалады.

Тірек сөздер: кремний нитриді, кремний оксиді, люминесценция, нанокластерлер, стехиометриялық тәртіп.

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ИССЛЕДОВАНИЕ РАЗЛИЧНЫХ МЕХАНИЗМОВ РАДИАЦИОННОЙ РЕКОМБИНАЦИИ НИТРИДА КРЕМНИЯ КАК ЭФФЕКТИВНОГО СПОСОБА РАСШИРЕНИЯ СПЕКТРА ФОТОЛЮМИНЕСЦЕНЦИИ

Аннотация

Разработка устройств, сочетающих оптические и электрические функции на основе кремнийсодержащих материалов, является одной из задач микроэлектроники. Путем плазменного синтеза с усиленным химическим осаждением из газовой фазы и последующего отжига были получены образцы нитрида кремния как с избытком кремния, так и с избытком азота. Высокую концентрацию связей Si-H и N-H определяли методом рамановской спектроскопии в образцах перед отжигом. С помощью просвечивающей электронной микроскопии было определено, что помимо нитрида кремния, в матрице образца образуются кластеры кремния. Спектры фотолюминесценции существенно изменялись для обоих типов образцов при отжиге в различных газовых атмосферах. Термическая обработка образцов при 1100 °C после синтеза привела к исчезновению спектра ПЛ, а после отжига при 800 °C фотолюминесценция увеличивается. Отмечено, что наибольшая интенсивность фотолюминесценции была обнаружена после отжига в воздушной атмосфере и наименьшая – в азоте. Участие N-центров в рекомбинационных процессах подтверждено методом электронного парамагнитного резонанса. Рассмотрены различные механизмы взаимодействия частиц, приводящих к фотолюминесценции и накоплению заряда. Таким образом, подобраны условия синтеза и отжига слоев нитрида кремния для получения управляемых люминесцентных свойств в различных спектральных диапазонах.

Ключевые слова: нитрид кремния, оксид кремния, люминесценция, нанокластеры, стехиометрический порядок.

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