Semiconductor Physics, Quantum Electronics & Optoelectronics, 2024. V. 27, No 3. P. 274-279.

Semiconductor physics

Redistribution of radiative recombination centers in the SiC/por-SiC/Dy_2O_3 structure under the influence of athermal microwave irradiation

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Abstract. In this work, the authors have considered the effect of short-term nonthermal action of microwave radiation on the distribution of radiative recombination centers in SiC/por-SiC/Dy_2O_3 structures. The analysis of photoluminescence spectra of these structures excited by the radiation with an energy lower than the band gap in the 4H-SiC crystalline substrate has shown that the short-term action of microwave radiation leads to the migration of dislocations and, as a consequence, to redistribution of radiative recombination centers and local symmetry change.

Keywords: athermal microwave action, rare-earth oxides, photoluminescence, silicon carbide.

https://doi.org/10.15407/spqeo27.03.274
PACS 81.07.-b, 85.40.Sz

Manuscript received 08.07.24; revised version received 24.08.24; accepted for publication 11.09.24; published online 20.09.24.

1. Introduction

Miniaturization of optoelectronic devices has necessitated the replacement of the traditional silicon oxide in metal-insulator-semiconductor (MIS) structures and integrated circuits (ICs) with alternative dielectrics (dielectrics with high dielectric constant – high-k dielectrics) [1–4]. Their use allows the increase of the physical thickness of the dielectric in the device and, thereby, suppresses the tunnel currents. Oxides of rare-earth elements (OREE) are promising dielectric materials for creating the insulating layers in the film structures of micro- and nanoelectronics. These materials have high thermal and chemical resistance, large (compared to silicon dioxide) dielectric constant values, and high electrical strength. The OREE films can be quite simply obtained on various semiconductor substrates at relatively low temperatures (300…800 °C), which is especially important for manufacturing the insulating layers on semiconductors that do not have good intrinsic thermal oxides. In addition, the OREEs have a high transparency within the spectral range 0.3…2.0 µm and have an optimal refractive index (n = 1.77…2.18) for their use as antireflective and passivating coatings in the photovoltaic devices [5]. The MIS varicaps, MIS transistors, and memory elements, based on some OREE dielectric films, with sufficiently high characteristics, heat-resistant, and effective antireflection coatings for photovoltaic devices have been developed [6–8]. However, the progress in the miniaturization of microelectronic devices causes a significant impact on the properties of the oxide-semiconductor interfaces on the parameters of MIS structures with thin dielectric films [1–4].

The additional external treatments, namely: temperature annealing, γ-irradiation, and microwave processing, are often used to modify the concentration of defect states at the oxide-semiconductor interface. The authors of [8] postulated that the perspective of using microwave treatments, to develop new technologies, is caused by the following factors. They are the possibility of implementing both short-term nano- and microsecond pulsed influence and heat treatment in a continuous mode; the possibility of selective homogeneous influence on the components of semiconductor device structures; and non-contact processing of materials in vacuum or special environments. Previously, we have observed the effect of changes in the absorption coefficient and redistribution of the intensity of photoluminescence bands for SiC/SiO_2 and SiC/TiO_2(Gd_2O_3, Er_2O_3), SiC/por-SiC/Er_2O_3 structures after short-term microwave irradiation [10–14]. These structures optical properties change due to the athermal exposure to microwaves [15]. According to the athermal interaction model [15], the resonant interaction of microwave radiation with dislocations of a certain length leads to the abruption of dislocations from stoppers and their movement both in the silicon carbide substrate and the oxide layer. In addition, when the frequency of microwave radiation is within the range of the stopper oscillations, the resonant abruption of dislocation from the stopper can be satisfied not only

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for dislocations of a strictly defined size but also for the dislocations of an arbitrary size but attached to a stopper oscillating at the resonant frequency.

Moreover, suppose we consider crystal lattice imperfections as dislocation stoppers. In that case, the abruption of dislocation from the stopper under the action of microwave radiation is also possible due to a decrease in the activation energy of mass transfer processes under the influence of the averaged ponderomotive force [16]. The movement of dislocation, in turn, leads to a change in the distribution of internal stresses in the structure and, consequently, to subsequent changes in the number and configuration of dislocations. Since the binding energy between an impurity atom and a dislocation is a function of the position of the defect related to the dislocation [16, 17], the movement of the dislocation caused by microwave irradiation should lead to a redistribution of the emission and absorption centers in the structures.

In this work, we investigated the effect of short-term athermal microwave irradiation on the distribution of radiative recombination centers in 4H-SiC/por-SiC/Dy_2O_3 structures.

2. Samples and measurement methods

A porous surface in silicon carbide (4H polytype) was obtained by electrochemical etching in a solution of HF:C_2H_5OH = 1:1 at the current density of 10 mA/cm^2, the etching time was 10 min. Then, the material was processed in a KOH melt at 550 °C to open the pores. At the next stage, an erbium film was deposited on the surface of porous silicon carbide by using the thermal sputtering method. Porous SiC structures with a deposited layer of dysprosium were annealed in vacuum at 800 °C for 8 min and then a thin oxide film of Dy_2O_3 was formed on the surface of por-SiC at a fast thermal annealing mode in atmosphere of dry oxygen at 400 °C. The fast thermal annealing time was 1…5 s. The thickness of the resulting oxide layers, determined by Auger spectroscopy [18], is ~100 nm.

Microwave processing was carried out in the magnetron operation chamber with the frequency f = 2.45 GHz and a specific power of ~ 0.04 W/cm^3. The microwave exposure time was 5 s. As was shown in [14, 19], the short-term microwave irradiation of 4H-SiC/por-SiC/Dy_2O_3 structures with the indicated parameters can be considered athermal.

Photoluminescence (PL) spectra of the samples were obtained in backscattering geometry by using a T64000 spectrometer (Horiba Jobin Yvon) with a confocal microscope and a cooled CCD detector.

The 532.0 nm line of the Ar-Kr laser was chosen to excite the PL attributed directly to radiative recombination in the por-SiC or por-SiC/Dy_2O_3 layer. This choice of the excitation wavelength is caused by the fact that the energy corresponding to λ_{ex} = 532.0 nm (2.33 eV) is significantly less than the band gap energy of the crystalline 4H-SiC (E_g =3.23 eV) [20]. All optical measurements were carried out at room temperature.

3. Experimental results and discussion

Fig. 1 shows the characteristic normalized PL spectra of SiC/por-SiC/Dy_2O_3 structures before and after microwave irradiation, excited with $h\nu_{ex}$ = 2.33 eV < E_g (4H-SiC) = = 3.23 eV. This choice of the photoluminescence excitation wavelength allows the registration of the PL spectrum caused by the radiative recombination that occurs only in the por-SiC/Dy_2O_3 layer. In this case, the 4H-SiC substrate (E_g = 3.23 eV) is transparent for this radiation.

It should be noted that a distinctive feature of por-SiC is the presence of photoluminescence even when the excitation radiation is less than the band gap energy of the initial crystalline material 4H-SiC and 6H-SiC substrate (E_g = 3.23 eV and E_g = 3.02 eV, respectively) [14, 21–23]. In this case, the PL spectra of por-SiC for the 4H-SiC, 6H-SiC (α-SiC) and 3C-SiC (β-SiC) polytypes have almost the same line shape [21, 22, 24].

The authors of [23] show that the appearance of PL in por-SiC obtained by anodic etching at the excitation with $h\nu_{ex} \le E_g$ is associated with the formation of radiative centers by impurity atoms and surface defects induced by the anodic etching of the crystalline SiC substrate and the corresponding processes that open the pores.

Thus, during the etching of the crystalline silicon carbide through the disruption of Si–C bonds in the por-SiC layer, the complex compounds, namely oxides, siloxenes, and Si–H or C–H bonds as well as C–N bonds form (nitrogen is an uncontrolled impurity in SiC) [21, 25–28]. Moreover, in the por-SiC an inessential presence of 3C-SiC phase as structural defects, namely stacking faults, can be.

As can be seen in Fig. 1, the PL spectrum of SiC/por-SiC/Dy_2O_3 structures is a broad complex peak, which consists of several overlapping bands centered at 590, 650, and 750 nm. These PL bands can also have a complex structure. The band at 590 nm is probably caused by the microcrystallites of cubic silicon carbide 3C-SiC in the por-SiC layer [29, 30]. The origin of the low-energy peaks at 650 and 750 nm can be explained by radiative recombination in donor-acceptor pairs [31] or by radiative transitions with the participation of oxygen-related defects [14].

After the microwave treatment of the SiC/por-SiC/Dy_2O_3 structure, a short-wavelength shift of the integral peak position and a change in the shape of the integral PL spectrum are observed, which correlates with previously obtained data for PL of the SiC/por-SiC/Er_2O_3 structures [14]. The noted shift is caused by the redistribution of the intensity of individual components constituting the integral PL contour. Thus, as can be seen in Fig. 1, after microwave treatment the intensity of the band at 590 nm increases, and the intensity of the bands at 650 and 750 nm decreases. Moreover, the intensity of the band at 750 nm is almost two-fold decreased. This decrease in the PL intensity can indicate the decrease in the number of radiative recombination centers associated with oxygen.

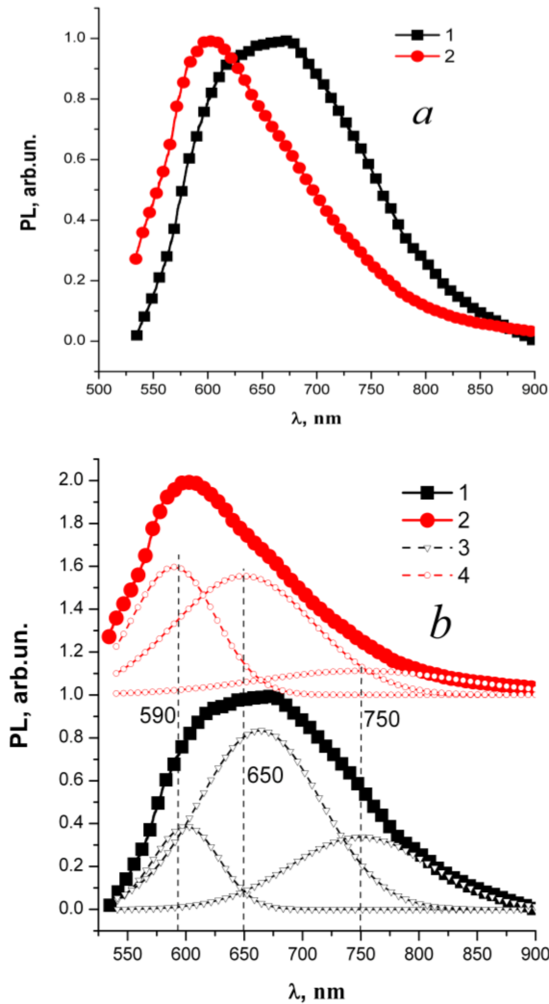

Fig. 1. Normalized PL spectra for the SiC/por-SiC/Dy_2O_3 structure *a*) before (*1*) and after (*2*) microwave irradiation and *b*) their decomposition on elementary components before (*3*) and after (*4*) microwave irradiation.

The integrated PL spectrum of the SiC/por-SiC/Dy_2O_3 structure, in contrast to the spectra of the SiC/por-SiC/Er_2O_3 structure, is a superposition of the PL spectra of por-SiC and Dy_2O_3 [32, 33].

With account that microwave treatment does not lead to a significant change in the ratio of the crystalline phases 4H-SiC/3C-SiC in the SiC/por-SiC and SiC/por-SiC/Er_2O_3 structures [14], it can be assumed that the observed blue shift of the maximum of the integral PL of the SiC/por-SiC/Dy_2O_3 induced by the microwave treatment is most likely associated with an increase in the intensity of the band at 577 nm attributed to the intracenter transitions in ion Dy^{3+} ($^4F_{9/2} \rightarrow {}^6H_{13/2}$) in the Dy_2O_3 [32, 33] film. A characteristic feature of this PL band of Dy^{3+} ion is the strong dependence of its intensity on the symmetry of the coordination environment [32, 33]. An increase in the intensity of the PL band at 577 nm dominates in the PL spectrum of Dy_2O_3 only when Dy^{3+} ions are located in low-symmetry sites without inversion centers [32, 33].

This hyper-dependence on the symmetry of the coordination environment allows, among other things, using this band intensity as a marker for determining the degree of distortion of the coordination environment of the Dy^{3+} ion.

The assumption that the symmetry of the nearest environment of the Dy^{3+} ion changes under the influence of microwaves correlates with the data of [11]. As it was noted in [11], microwave irradiation of the SiC/SiO_2 structures leads to fluctuations in the inhomogeneity of distribution of dopant impurities and defects both on the surface and in the bulk of the structure, resulting in a change in the nature of the inter-impurity interaction between PL and absorption centers. Thus, the authors of [11] observed the change in the local symmetry for nonequivalent nitrogen donors with hexagonal and cubic coordination of the nearest environment in the SiC/SiO_2 structures under the influence of microwave treatment, which led to the redistribution of intensities of individual bands constituting the integral absorption profile of the SiC/SiO_2 structure [11].

4. Conclusions

Thus, the athermal effect of microwave radiation on the SiC/por-SiC/Dy_2O_3 structures manifests in the redistribution of radiative recombination centers in these structures. This redistribution of PL centers can be caused by the abruption of the dislocation from the stoppers under the influence of microwave irradiation, which leads to their further migration within the field of mechanical stresses of the crystal lattice. Consequently, the movement of dislocations caused by microwave irradiation leads to the redistribution of radiative centers and a change in their local symmetry in the SiC/por-SiC/Dy_2O_3 structures.

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Перерозподіл центрів випромінювальної рекомбінації в структурі SiC/por-SiC/Dy_2O_3 під впливом атермічної СВЧ дії

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Анотація. У даній роботі розглянуто вплив короткочасної нетеплової дії мікрохвильового випромінювання на розподіл центрів випромінювальної рекомбінації в структурах SiC/por-SiC/Dy_2O_3. Аналіз спектрів фотолюмінесценції цих структур при збудженні випромінюванням з енергією, меншою за ширину забороненої зони кристалічної підкладки 4H-SiC, показав, що короткочасна дія мікрохвильового випромінювання приводить до міграції дислокацій і, як наслідок, до перерозподілу центрів випромінювальної рекомбінації та зміни їх локальної симетрії.

Ключові слова: нетеплова мікрохвильова дія, рідкісноземельні оксиди, фотолюмінесценція, карбід кремнію.