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Cathodoluminescence and epitaxy after sequential Rb- and Ge-ion implantation in α-quartz

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Abstract

The luminescence properties of ion-beam doped silica and quartz sensitively depend on the ion species and fluence and the thermal processing during or after ion implantation. While no epitaxy, but a high cathodoluminescence (CL) output can be reached after dynamic Ge-ion implantation in quartz, full planar epitaxy is achieved after alkali-ion implantation (Na, Rb, Cs) under appropriate ion and annealing conditions. Now for the first time we achieved high luminescence intensity and (partial) recrystallization of α-quartz by double Ge/Rb-ion implantation. Pure synthetic α-quartz samples were irradiated with 175 keV Rb-ions and subsequently with 120 keV Ge-ions and post-annealed at 1170 K in vacuo, air or $^{18}O_2$ gas. A comparative analysis of the epitaxy (via RBS-Channeling) and cathodoluminescence was carried out, the latter showing three dominant emission bands in the blue/violet region at 2.95, 3.25 and 3.53 eV, which were assigned to Rb- or Ge-implantation. For increasing CL temperature, a shift of the predominant peak from 2.95 to 3.25 eV was noted after air-annealing, accompanying partial chemical epitaxy.

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Keywords: Ion implantation; Soild phase epitaxy; Cathodoluminescence; Quartz

1. Introduction

The role of photoactive defects and nanoparticles in crystalline and amorphous silicon dioxide used to tune its luminescence properties has gained renewed interest in photonic technology. Ion implantation and subsequent annealing is a suitable way to produce such defects in silica and quartz for fabricating various optoelectronic and photonic devices such as optical insulators, waveguides, and switches [1–3]. In the last two decades, many studies have been devoted to dope a-SiO_2 with semiconductor or rare-earth element ions, using various processing techniques. Photoluminescence (PL), cathodoluminescence (CL) and

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infrared (IR) absorption spectroscopy are the leading techniques to investigate the emission and absorption bands, which are fairly broad and often overlap. Silica emits strong blue luminescence when exposed to ultraviolet light [4,5]. Definite correlations were found between some of the optical absorption and emission bands and specific defect structures, but important information on their properties, such as the size of nanoparticles, the lattice location, oxidation and charge state of the implanted ions, and the role of vacancy trapping, are still lacking. Kalceff [6] and Skuja [7] recently reviewed the CL and PL emission bands of ion-irradiated silica. Skuja and Entzian [8] assigned the CL bands at 2.7 and 4.3 eV to the triplet and singlet luminescence of the twofold-coordinated silicon centers. Implantation of Ge- or Si-ions into a-SiO_2 followed by high-temperature annealing produces strong luminescence in the visible range (violet and blue bands) [9,10].

As an alternative, modifying the optical properties of single-crystalline synthetic α-quartz by ion implantation is possibly advantageous for various applications, although quartz easily becomes amorphous during ion implantation [11,12] and usually does not recrystallize under thermal treatment. Hence, simultaneously achieving high light emission intensity by appropriate doping and good epitaxial recovery of quartz is a challenge. In the past, several attempts have been undertaken to achieve epitaxy of ion-irradiated quartz via dynamic annealing [13], laser irradiation [14] or chemical epitaxy [15–18]. The latter process uses alkali-ion implantation (Na, Rb, Cs) followed by thermal annealing in air or oxygen. Almost complete recrystallization has been also achieved by dynamic epitaxy after Ba-ion implantation [19].

The present work is based on our recent results concerning the cathodoluminescence after Rb- or Ge-ion implantation and dynamic or chemical epitaxy in quartz [17,18,20]. Ge-implantation leads to intense blue and violet CL bands, but does not produce full dynamic epitaxy up to 1170 K [20]. On the other hand, Rb-ion implantation leads to full chemical epitaxy upon thermal treatment in air or oxygen, but the implanted ions diffuse out and the CL light output becomes rather weak [17,18]. These findings suggested that Ge and Rb double-ion implantation followed by an appropriate annealing in oxygen may be appropriate to retain the strong Ge-related CL bands and at the same time recrystallize the SiO_2 matrix. The objective of the present investigation was to realize this idea and to optimize the processing parameters in order to gain both epitaxy and maximum light output.

2. Experiments

One-side polished samples of synthetic α-quartz, (0001) oriented and $10 \times 10\ mm^2$ in size, were irradiated with 175-keV Rb^+-ions at liquid nitrogen temperature at a fixed fluence of 2.5×10^{16} ions/cm^2. Subsequently the samples were irradiated with 120-keV Ge^+-ions at room temperature at fluences of up to 1×10^{16} ions/cm^2. Homogeneous implantations with a beam flux of 3×10^{12} ions/cm^2 s was maintained for all the samples. After the double implantation each sample was annealed at 1170 K for 1 h in air, $^{18}O_2$ or vacuo. The damage analysis and the Ge and Rb depth profiles were monitored by means of Rutherford Backscattering Channeling Spectroscopy (RBS-C) with a 0.9 MeV He^{++} beam. The detailed experimental conditions and analyses are described in [20].

The cathodoluminescence spectra were recorded in the temperature range from 10 to 300 K before and after annealing. The samples were mounted on an oxygen-free copper target head of a closed-cycle helium cryostat and irradiated with a 5-keV electron beam of 1 W/cm^2 power density (Specs EQ-22). The CL light was collected using an achromatic lens and monitored by means of a photo-multiplier tube (Hamamatsu R928). Each CL spectrum was collected in the wavelength range of 200–800 nm using a 1200 lines/mm grating.

3. Results and discussion

3.1. Solid phase epitaxy

Fig. 1 shows the RBS-channeling spectra of 120-keV Ge^+ implantation, with or without previ-