Luminescence in GeO_x Films Containing Germanium Nanoclusters

K. N. Astankova^a, E. B. Gorokhov^a, V. A. Volodin^{a, b}, D. V. Marin^a, I. A. Azarov^a, and A. V. Latyshev^{a, b}

 ^aRzhanov Institute of Semiconductor Physics, Siberian Branch, Russian Academy of Sciences, pr. Akad. Lavrentieva 13, Novosibirsk, 630090 Russia
^bNovosibirsk State University, ul. Pirogova 2, Novosibirsk, 630090 Russia
e-mail: as-tankoff@va.ru

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Abstract—Metastable GeO_x films have been deposited onto a Si substrate by the electron beam evaporation of GeO₂ powder in high vacuum. The optical properties of Ge nanoclusters in GeO_x films after a series of annealing are studied by Raman spectroscopy, ellipsometry, cathodoluminescence, and photoluminescence. After the annealings, the cathodoluminescence peaks are first found in GeO_x films in the visible spectral range (400 and 660 nm) at room temperature. The cathodoluminescence may be associated both with the existence of nonground (excited) levels in Ge nanoclusters and allowed optical transitions from an excited to ground level and surface states at Ge/GeO₂-matrix interface or defects inside a Ge nanocluster. The photoluminescence signals in a yellow-green region (2.1–2.4 eV), which are observed for GeO_x films after annealings, can be explained by quasi-direct optical transitions in Ge nanoclusters.

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INTRODUCTION

The ensembles of semiconductor nanoparticles (quantum dots) incorporated in a wide-bandgap dielectric matrix attract great attention from both the fundamental point of view and their possible practical application in nanoelectronics and nanophotonics. One important circumstance is the fact that, due to the quantum size effect, a radiative recombination is observed in nanoclusters of indirect bandgap semiconductors even at room temperature, which is not inherent to them [1].

Over the last several years, structures with quantum dots have begun being applied for the production of solar cells, light-emitting diodes, sensors, displays, and biomarkers for medicine. Compared to the well-studied semiconductor quantum dots based on A_{II}-B_{VI}, A_{IV}-B_{VI}, and A_{III}-B_V compounds (e.g., CdSe, PbS, PbSe, PbTe, InAs, InP, etc.) in colloid solutions [2] or in a solid matrix [3]. Ge nanoclusters (Ge NC) in a dielectric are at a stage of intense studies [4, 5]. Moreover, more and more often the possibility of replacing toxic semiconductor materials with heavy metal compounds by Si and Ge in devices of mass consumption have began to be considered [6]. A comparison of IV-group semiconductors (Si and Ge) shows that, in certain applications, germanium possesses a number of advantages over silicon [7]. It has the highest hole mobility (1900 cm^2/V s) of all the semiconductors of the IVth group and the compounds of the III–V groups of the periodic table (GaAs, InSb, InP). The quantum size effect should be exhibited more brightly in germanium nanocrystals than in silicon nanocrystals, because the exciton radius in bulk germanium is several times larger than that in bulk silicon.

The appearance of light radiation in ultraviolet, visible, and infrared spectral regions under electron irradiation (cathodoluminescence) has been known a long time. The method of cathodoluminescence (CL), together with other methods such as photoluminescence (PL) and Raman spectroscopy (RS), are widely used in research by studying the energy-band structure of solids (mainly of those energy levels, which participate in the processes of radiative recombination).

EXPERIMENTAL TECHNIQUE

Films of solid germanium monoxide (GeO(sol)) are metastable and easily decompose into the GeO_2 matrix with Ge NCs included into it according to the following chemical reaction [8]:

 $2\text{GeO}(\text{sol}) \rightarrow \text{Ge}(\text{sol}) + \text{GeO}_2(\text{sol}).$

Depending on the growth conditions, it is possible to obtain either homogeneous GeO films of a stoichiometric composition [9, 10] or GeO_x films of various compositions and structures [11, 12, 13]. In this work we used the method of electron beam evaporation of GeO₂ powder in a high vacuum chamber (10^{-8} Tor). Under the action of the electron beam, a partial decomposition of GeO₂ into Ge, O₂, and GeO takes place. The latter two components are more volatile,