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Luminescent properties of ordered arrays of silicon disk-like resonators with embedded GeSi quantum dots

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The emission properties of ordered arrays of silicon disk-like resonators with embedded GeSi quantum dots are studied. It is shown that, depending on the distance between the resonators, the structures can exhibit the properties of isolated Mie resonators or photonic crystals characterized by the presence of a contribution from photonic crystal modes in the photoluminescence spectrum. The formation of photonic crystals based on the disk-like resonators makes it possible to significantly increase the luminescence response in the wavelength range of 1.2−1.6 μm, even at room temperature.

Keywords: GeSi quantum dots, photonic crystals, Mie resonances, luminescence.

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1. Introduction

Currently, dielectric resonators are widely used to control the optical properties of various structures, the distinctive feature of which is low losses [1–3]. One of the most relevant areas of use of dielectric resonators is the modification with their help of the radiative properties of the active medium built into the resonator [4–9]. At the same time, depending on the specific problem being solved, both individual resonators [4,5] and their arrays as metasurfaces are considered [1,3,6,7]. Work in this direction led to the creation of $\mathrm{A^{III}B^{V}}$ family of lasers on single disk resonators [8] and arrays of such resonators [9,10] on straight-band materials.

However, it seems promising from the point of view of compatibility with integrated silicon technology and the development of silicon optoelectronics to use dielectric resonators to improve the radiative properties of structures based on silicon and its alloys with germanium. This, in particular, applies to structures with GeSi self-forming nanostructures and quantum dots (QDs), demonstrating at room temperature a luminescence signal in the wavelength range from 1.2 to 1.6 μm [11–14], used in silicon optoelectronic devices and fiber-optic communication systems [14]. These structures are relatively easy to form, compact and compatible with modern integrated silicon technology [12,14]. These qualities made structures with GeSi QD convenient for studying the interaction of the active medium with the modes of various dielectric resonators, such as the modes of microresonators in two-dimensional photonic crystals (FC) [15,16], the modes of the FC themselves [17–21], as well as the modes of single and linear chains of disk resonators [5,22]. Interaction with resonator modes led to an increase in the integral intensity of the luminescence signal and the appearance of narrow peaks in the emission spectra of structures with GeSi QD due to the interaction of QD with high-quality resonator modes [5,7,15–22].

Works dedicated to the interaction of GeSi QD with different dielectric resonators can be conditionally divided into two groups. The interaction of QD with modes of a single resonator [5] or an ensemble of their small number [5,22] is considered in one of them. The other includes the study of FC [15–21] and metasurfaces [7], which can be considered as an array of a large number of interacting resonators. In this case, the interaction of GeSi QD occurs with collective modes of the resonator array. At the same time, the transition from the GeSi QD interaction with modes of a single resonator to their interaction with collective modes of an array of interacting resonators with an increase in the number of QD or a decrease of the distance between them remains insufficiently studied.

This paper presents the results of the formation of single and arrays of disk resonators with Ge(Si) quantum dots and studies of their luminescent properties. Depending on the distance between the resonators, the transition from the interaction of QD with the modes of single resonators to the interaction with high-quality collective modes of FC formed by a square lattice of disk resonators is demonstrated.

2. Experiment procedure

Arrays of disk resonators were formed on the basis of multilayer structures with GeSi QD grown by molecular beam epitaxy on „silicon-on-insulator“ substrates (SOI). The thickness of the upper silicon layer in the SOI substrates was 170 nm, and the hidden layer SiO_2 — 3 μm. Previously, a buffer layer of SI with a thickness of 110nm was grown on the surface of the SOI substrate with a gradual increase in the growth temperature from 250 to 500°C, followed by annealing at 700°C for 10 min. Next, a multilayer structure containing 10 layers of GeSi QD was grown (each QD layer was formed by deposition of ~ 1 nm germanium) alternating with Si layers with a thickness of 15 nm. Small thickness of Si layers separating adjacent layers with GeSi QD ensured vertical alignment of QD. Growth temperature of GeSi QD, intermediate Si layers, as well as the Si cover layer with a thickness of 20 nm was 600°C. According to ellipsometry data, the total thickness of the structure above the layer SiO_2 was ~ 450 nm.

The disk resonators were formed in several stages. At the first stage, a mask was created using electron beam lithography in a positive resistor PMMA-950K with a thickness of ~ 200 nm, representing arrays of round holes located in the nodes of a square lattice with a period from 1 μm to 6 μm. The radius of the holes in the resistance mask varied in the range from 300 ± 10 nm to 360 ± 10 nm in increments of 20 nm. Next, a thin layer of chromium (~ 20 nm) was deposited on top of the mask with holes by magnetron sputtering. Periodic arrays of Cr disks were obtained after removing the resist on the surface of the multilayer structure with GeSi QD, which served as a mask for creating resonators. At the next stage, anisotropic plasmochemical etching of Ge/Si structures in a mixture of gases $SF_6:SHF_3$ to a hidden layer SiO_2 was carried out through the Cr mask. Chromium was removed from the surface of the structure by liquid etching at the last stage.

Thus, arrays of disk resonators were created in the form of silicon nanocylinders with radii ranging from 320 ± 10 to 390 ± 10 nm, located at the nodes of square gratings with a period from 1 to 6 μm (see details in Figure 1). According to the study of the obtained disk resonators using scanning electron microscopy (SEM), they had vertical side walls with low roughness, and their height ~ 450 nm corresponded to the total thickness of the structure above the layer of latent oxide. The photoluminescence (PL) signal of the resonators was compared with the signal from the original structure. Rectangular areas $100 \times 30\,\mu$m, were created on the sample to do this, which were not subjected to plasma chemical etching (unprocessed areas). They, like disk resonators, had vertical side walls.

Luminescent measurements of the obtained disk resonators were performed using the standard microphotoluminescence (micro-PL) technique in the geometry of the normal incidence of exciting radiation and the detected signal [20]. The PL signal was excited by a solid-state laser at a wavelength of 532 nm. Mitutoyo M Plan APO NIR lens with a 50x magnification (numerical aperture $NA = 0.42$) was used in the study for precision focusing of the laser beam on the sample. The lens provided the ability to focus the laser beam into a spot with a diameter of $\sim 2\,\mu$m. Spectral measurements with a resolution of 4 cm^{-1} were performed using spectrometer Bruker IF 125 HR Fourier. The PL signal was recorded using a cooled Ge-photodetector. The measurements were carried out at room temperature and liquid nitrogen temperature (77 K) in a flow cryostat.

Theoretical calculations of the band diagrams of photonic crystals represented by square arrays of disk resonators were performed by the Fourier modal method in the approximation of the scattering matrix [23].

3. Results and discussion

Figure 1 shows the spectra of microfl structures with resonators arranged in square lattices with periods of 1 μm (*a*) and 2 μm (*b*), depending on their radius. Measurements were made at temperature of 77 K. As can be seen from the figure, as the radius of the resonators increases, the spectral position and intensity of the PL peaks change in the energy range from 750 to 1050 MeV. In this case, the effect of amplification of the signal intensity is greater, the larger the radius of the resonators. A structure with a period of 1 μm demonstrates narrow intense peaks in the micro-PL spectra, both in the region of GeSi QD radiation, and in the radiation region of the Ge wetting layer (Figure 1, *a*) [24]. Only widened peaks of PL with a width of ~ 50 MeV are observed in arrays of resonators with a period of 2 μm (Figure 1, *b*). In both cases, the spectral position of the intensity maxima depends on the radius of the resonators and shifts, as the radius increases, to the region of lower photon energies. The latter indicates the relationship of the observed signals with the radiative modes of the studied resonators. It should be noted that a PL signal associated with a silicon substrate is observed in the PL spectra of the studied resonator arrays at a temperature of 77 K (peak Si_{TO} in Figure 1, *b*). The intensity of this signal depends on the conditions of focusing the laser beam on the disk resonator and increases as the size and density of the resonators in the structure decrease.

The limiting case of arrays of disk resonators with long periods, when the interaction between the resonators can be neglected, are arrays of resonators with a period of 6 μm. The micro-PL spectra of such arrays show wide peaks, the

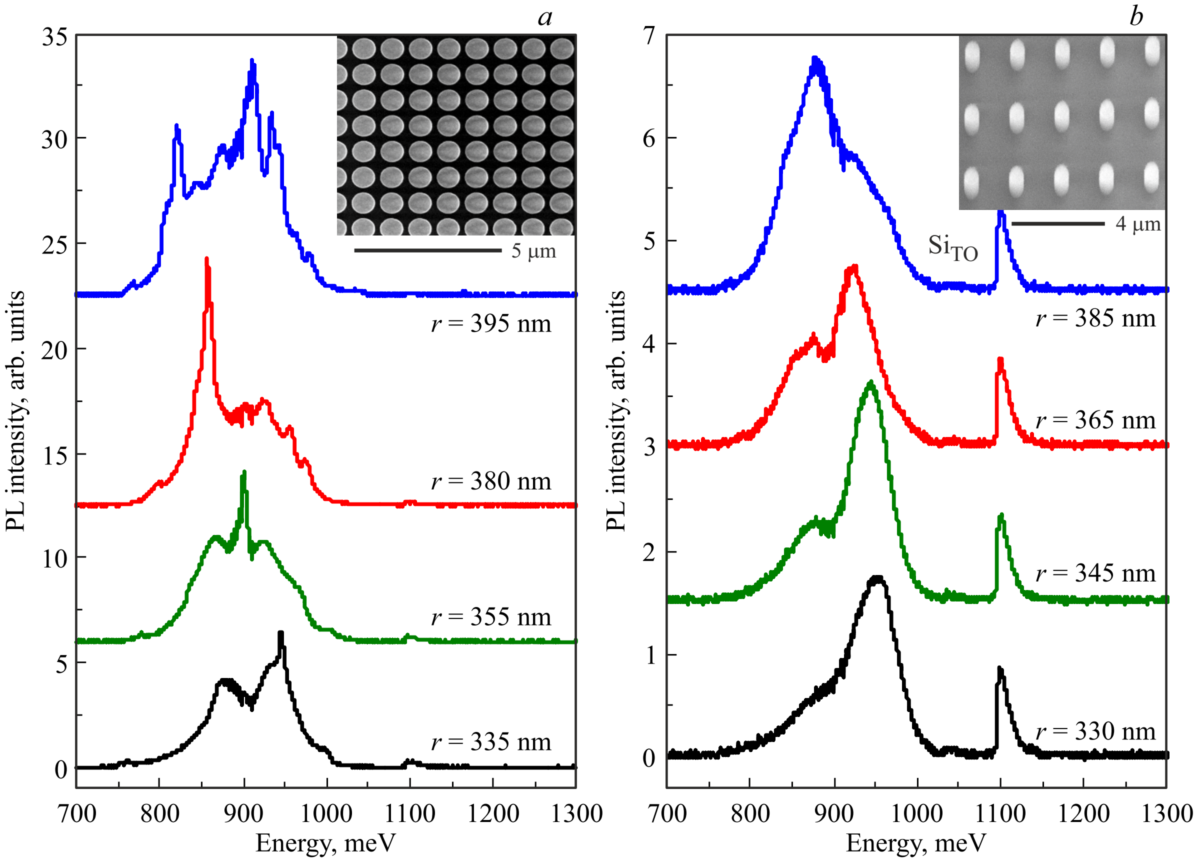

Figure 1. The spectra of micro-PL structures with disk resonators arranged in square lattices measured at a temperature of 77 K, the periods of square lattices: 1 μm (a) and 2 μm (b). The radii of the disk resonators obtained from the SEM data are shown near the corresponding spectrum. Peak „Si_{TO}“ in the figure — PL signal from a silicon substrate. The inserts show SEM images of disk resonators arranged in square lattices with corresponding periods: a — lattice period 1 μm, radius of disks 385 ± 10 nm; b — lattice period 2 μm, the radius of the disks 320 ± 10 nm. The spectra are shifted vertically for clarity. (A color version of the figure is provided in the online version of the paper).

position of which shifts to the region of low photon energies as the radius of the resonator increases. The distance between the resonators in this case significantly exceeds both their own size and the characteristic wavelength of the radiation at which the signal is observed. Therefore, resonators can be considered as single, non-interconnected objects. In this case, the observed peaks in the PL spectra are naturally associated with the resonances occurring in such structures [5]. Earlier resonances of Mi in structures with GeSi QD scans were observed in silicon cylindrical columns with diameters of 280–660 nm. The authors of the work [5] showed that the resonances of Mi at energies close to 900 MeV have the character of an electric quadrupole with a resonator diameter of 600 nm and as the size of the resonator increases, the resonance position shifts to the region of lower energies. A similar shift in the spectral position of the peaks is observed in our experiment.

As the distance between the resonators decreases from 6 to 2 μm, an increase in the PL signal is observed, while the type of spectra practically does not change. It can be assumed that the growth of the PL signal observed in arrays with periods of 6, 4, 3 and 2 μm is due to the processes of scattering of laser radiation on the resonator array and the involvement of an increasing number of resonators in the signal, which leads to its growth. Thus, it is possible to distinguish the case of single resonators in the studied arrays of disk resonators with different radii and the distance between them that do not interact with each other, in whose PL spectra the resonance features of Mi are manifested. This case for the studied resonators is realized at distances between them in the range from 2 to 6 μm.

Cardinal changes are observed in the spectra of micro-PL arrays of disk resonators with a distance between the resonators of 1 μm (Figure 1). Along with the increase in signal intensity, narrow lines appear in the PL spectra of such arrays, the spectral position of which depends on the radius of the resonator (Figure 1, a). A sharp change in the spectra and the appearance of narrow lines of high Q-factor in the spectra indicate a change in the nature of the observed resonant phenomena. A possible explanation here is that at periods of 1 μm, the distance between the resonators is small and the square lattice of disk resonators

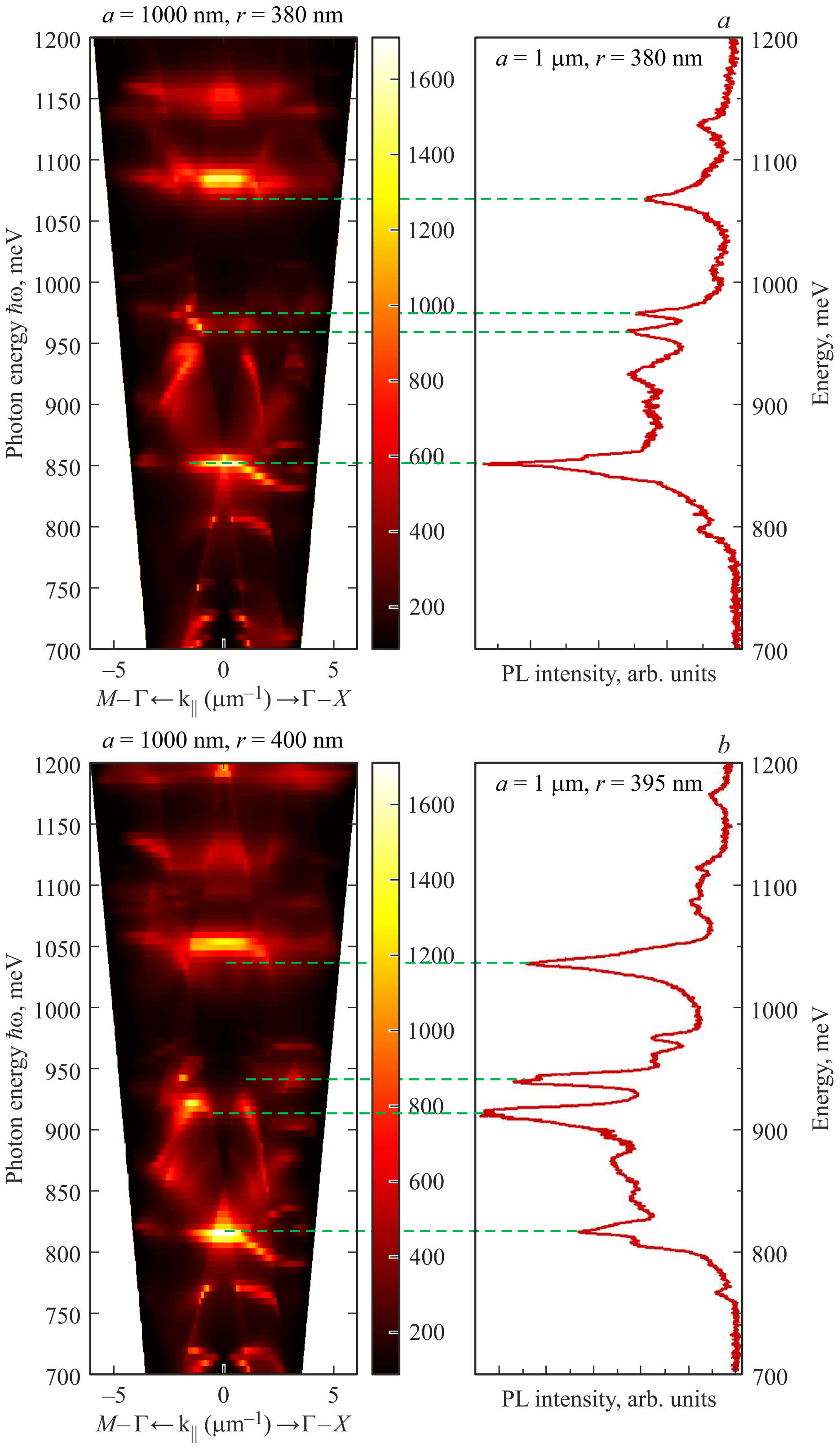

Figure 2. Calculated dispersion dependences of the far-field emissivity for FC represented by square arrays of cylindrical silicon columns with a period of 1 μm, with radii: 380 (*a*) and 400 nm (*b*). On the right, figures *a* and *b* show the spectra of micro-PL structures with square arrays of disk resonators, the parameters of which are close to the parameters used in the calculations. The dotted lines in the figures show the correspondence of the observed maxima in the micro-PL spectra with the FC modes. (A color version of the figure is provided in the online version of the paper).

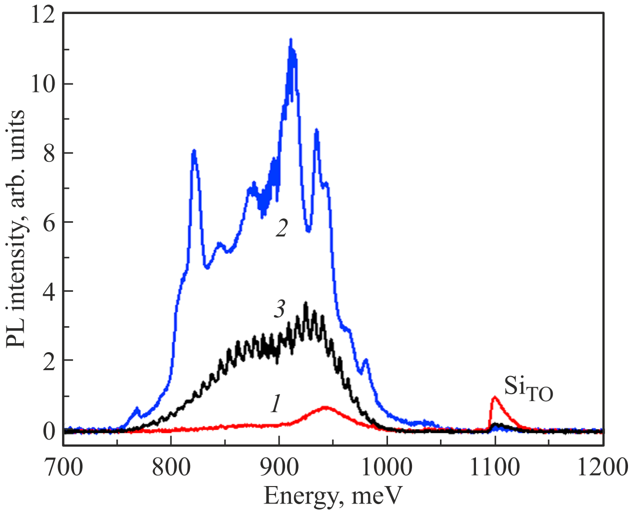

Figure 3. Micro-PPL spectra: of a single disk resonator with a radius of 365 ± 10 nm (*1*), an array of disk resonators with a radius of 395 ± 10 nm, ordered in a square lattice with a period of 1 μm (*2*) and the unprocessed region of the sample (*3*). The spectra were measured at a temperature of 77 K. (A color version of the figure is provided in the online version of the paper).

we are considering functions as a photonic crystal. Indeed, photonic crystals can be realized both on the basis of holes formed in a semiconductor material, and columns of the semiconductor material itself, ordered in a square or hexagonal lattice [6,25].

Theoretical calculations of the band structure of photonic crystals with parameters corresponding to the parameters of the studied arrays of disk resonators were carried out. Figures 2, *a* and *b* show the results of calculations of the dispersion dependences of the emissivity in the far field of photonic modes of structures with cylindrical columns of silicon arranged in a square lattice with a period of 1 μm, with radii of columns 380 and 400 nm, respectively. The same figures on the right show the measured micro-PL spectra of square arrays of disk resonators. The spectra are combined in energy with the data of theoretical calculations. It can be seen from Figure 2 that the position of the narrow peaks in the PL spectra agrees well with the calculated energy position of the FC modes. This indicates that there is an interaction of the emitting medium (quantum dots, wetting layer) with the modes of the photonic crystal. Thus, in the case of small periods (small distance between neighboring resonators), ordered arrays of disk resonators function as photonic crystals, demonstrating an intense PL signal, the spectrum of which is determined by the band structure FC.

Let’s compare the luminescent response from the obtained arrays of disk resonators with the signal observed in the initial, unprocessed region of the sample (Figure 3). Immediately note that the signal from the QD in the unprocessed regions is modulated with a period of 6–8 MeV (curve *3*). The modulation of the PL signal is apparently related to Fabry-Perot resonances occurring on the side walls of the regions. As can be seen from the figure, at $T = 77$ K is the signal of the PL array with a period of 1 μm, the spectrum of which is controlled by the interaction of GeSi QD with FC modes (curve *2*) significantly exceeds the signal observed in the unprocessed region of the sample (curve *3*). Thus, the increase in the intensity of the PL peaks at energies 820 and 910 MeV with respect to the signal at these energies in the unprocessed region of the sample is, respectively, ~ 6.5 and ~ 3.4 times. At the same time, the integral intensity of the PL signal from a structure with arrays of resonators with a period of 1 μm in the energy range from 750 to 1050 MeV exceeds the integral intensity of the PL signal in the unprocessed region by ~ 3 times. For single resonators (an array of resonators with a period of 6 μm, curve *1*), the situation is reversed. Here, the resonator’s PL signal is at a maximum 4 times smaller than the signal measured at the same energy value in the unprocessed region of the sample. The integral intensity of the PL signal of a single resonator is 8 times less than the integral intensity of the signal measured in the unprocessed region. This roughly corresponds to the ratio of the radiating volumes of the resonator and the unprocessed region. The real size of the resonator and the size of the excited unprocessed region of the sample were used to estimate the ratio of radiating volumes. The diameter of the focused laser beam was ~ 2 μm.

Arrays of disk resonators located with a period of 1 μm, with GeSi QD embedded in them is also radiated quite effectively at room temperature. Figure 4 shows the micro-PL spectra of an array of disk resonators with radii of 395 nm and a period of 1 μm, measured at temperatures of 77 and 300 K. The intensity of the PL signal of this array decreases with an increase in temperature from 77 to 300 K in ~ 13.5 times (the ratio of intensities in the maximum of the signal). At the same time, the PL signal remains quite intense and well detectable at room temperature. However, it should be noted here that the PL signal measured in the unprocessed region of the sample decreases with an increase in temperature from 77 to 300 K only 4.2 times, which is significantly less than the drop in signal intensity in the resonator (the signal from the unprocessed region at 300K, as in the case of 77 K, modulated due to Fabry–Perot resonances on the side walls of the region). A more pronounced decrease in the intensity of the PL signal in the resonator with an increase in temperature is apparently associated with an increase in the contribution of non-radiative recombination channels to photoluminescence quenching due to a more developed resonator surface compared to the unprocessed region. As a result of different temperature attenuation of the intensity of the PL signals from arrays of disk resonators with a period of 1 μm and the unprocessed region of the sample, measured at room temperature, are comparable (Figure 4). However, in the PL spectrum from an array of resonators with a period of 1 μm at room temperature, modes localized in the range of radiation energies of the

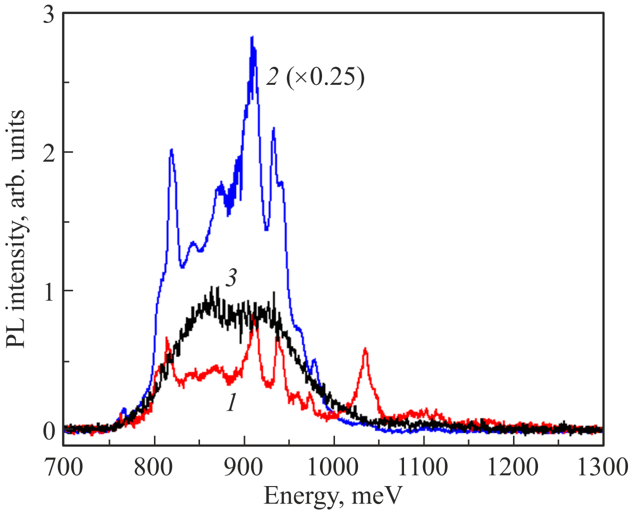

Figure 4. Micro-PL spectra of an ordered array of resonators with disk radii 395 ± 10 nm and a period of 1 μm, measured at temperatures 300 K (*1*) and 77 K (*2*), as well as the spectrum of micro-PL the unprocessed area of the sample, measured at 300 K (*3*). For clarity, the micro-PL spectrum of the disk resonator array, measured at a temperature of 77 K, is multiplied by 0.25. (A color version of the figure is provided in the online version of the paper).

wetting layer of the structure under study begin to manifest (see energy range $\sim$ 1035 MeV in Figure 4), the intensity of which is 7 times higher than the intensity the PL signal from an unprocessed region in this energy region. Thus, the choice of the parameters of the resonator lattice and their sizes makes it possible to control the radiating properties of such structures within a wide range, controlling not only the intensity, but also their spectral response.

4. Conclusion

Luminescent properties of arrays of disk resonators with GeSi QD ordered in a square lattice with periods from 1 to 6 μm, with resonator radii varying from 320 to 395 nm are studied in this paper. It is shown that for lattice periods in the range from 2 to 6 μm, disk resonators manifest themselves as single resonators with a characteristic radiation spectrum in which the position of the peaks of the PL depends only on the size of the resonator. An increase in the intensity of the PL signal is observed with a decrease in the period from 6 to 2 μm in arrays of resonators arranged in a square lattice, explained by the scattering processes of exciting laser radiation in a dense array of resonators and, as a consequence, an increase in the number of resonators contributing to the detected PL signal. Arrays of disk resonators arranged in a square lattice represent a photonic crystal with lattice periods of 1 μm, and the fine structure of PL signal lines observed in such arrays agrees well with the features of the band structure of a photonic crystal calculated theoretically. The intensity of the GeSi QD PL signal increases in the studied resonator arrays, which can be associated with the interaction of QD with both Mi resonances and collective FC modes. For certain FC modes, the PL signal increased 6.5 times at a temperature of 77 K and 7 times at a temperature of 300 K. The structures considered are of interest from the point of view of the possibility of creating near-infrared radiation sources based on them for silicon integrated circuits with optical signal processing.

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Conflict of interest

The authors declare that they have no conflict of interest.

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