Effect of optical excitation conditions on the spectral and temporal characteristics of the radiation from two-dimensional photonic crystals with Ge(Si) nanoislands

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The effect of optical excitation conditions on the spectral and temporal characteristics of the radiation from two-dimensional photonic crystals based on the structures with self-assembled Ge(Si) nanoislands has been studied. It was shown that one of the main factors effecting the spectral position and shape of photoluminescence lines, as well as the photoluminescence kinetics of photonic crystals with Ge(Si) nanoislands, along with the mode structure of the photonic crystal, are the local heating of the samples and the concentration of nonequilibrium charge carriers created by the absorption of the pumping radiation.

Keywords: self-assembled Ge(Si) nanoislands, two-dimensional photonic crystals, micro-PL spectroscopy, time-resolved PL studies.

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1. Introduction

At present, one of the actual directions of research in photonics is the modification of the radiation interaction with matter by creating microresonators and two-dimensional photonic crystals (PC) [1–5]. Of particular interest is the use of such microresonators and PCs to obtain efficient sources of near-infrared radiation based on SiGe structures, including structures with self-forming Ge(Si)-nanostructures, since such structures are part of modern silicon integrated technology [6,7] and demonstrate at room temperature a photo and electroluminescence signal in the spectral area $1.3-1.6\,\mu$m [8–12] used for information transfer in optoelectronic circuits and fiber-optic communication links. Structures with Ge(Si)-islands interstitial in photonic crystals and PC microresonators are the subject of intensive research aimed at controlling the optical properties of semiconductor materials [13,14]. Among these papers, a considerable amount is devoted to the study of Ge(Si)-islands in two-dimensional PCs that do not contain a microresonator [15–18]. The obvious advantage of such structures in comparison with structures with PC microresonators is the absence of a limitation on the size of the radiating area, as well as a narrow directivity diagram and the possibility of controlling the radiation directivity. The increase in the emission intensity of Ge(Si)-islands in such PCs can be realized both by increasing the efficiency of radiation output from the structure and by increasing the probability of radiative recombination of charge carriers due to the interaction of the active medium with the PC modes (the Parcell [19] effect).

In the present paper, structures with Ge(Si)-nano-islands interstitial in two-dimensional PCs were studied, in which a significant increase in the intensity of photoluminescence (PL) islands at certain wavelengths corresponding to the PC [17,18,20] modes and depending on the PC parameters was observed. It has been shown previously that the intensity and appearance of the PL spectrum of photonic crystals can be varied by varying the etching depth of the holes forming the PC [21]. In particular, it has been shown that at certain parameters of PC (period, diameter, and depth of hole etching), extremely narrow PL lines (width < 1 meV) are observed in the PL spectra of PC, the occurrence of which is associated with the presence of the so-called „bound-state-in-continuum“ (BIC) states in the energy band diagram of the PC, as well as with the formation of the „PLat gap“ in the PC band diagram, characterized by an extremely weak dependence of the

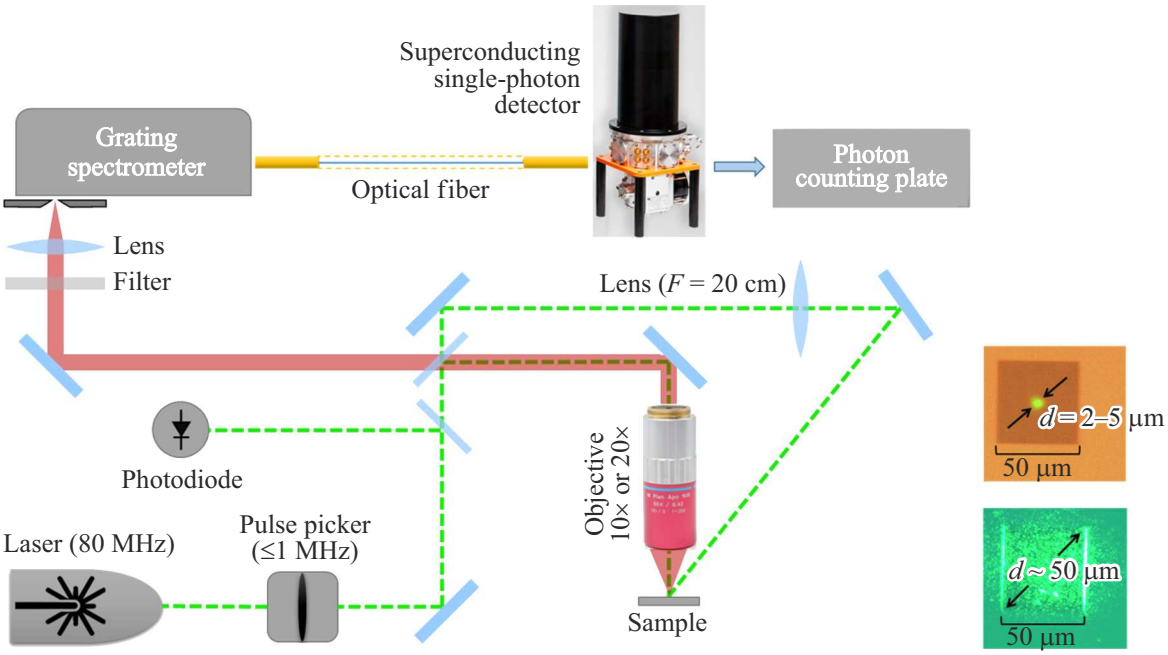

Figure 1. Scheme for measuring spectro-kinetic dependences of micro-PL with two possible variants of laser radiation focusing. Insert shows the image of the PC and excitation spot for two focusing options (*1* — $d \sim 2-5\,\mu$m, *2* — $d \sim 50\,\mu$m). (A color version of the figure is provided in the online version of the paper).

photon energy on the exit angle of radiation from the structure [22].

Along with the conditions for obtaining the initial structures with Ge(Si)-nanostructures and with the parameters of the PCs formed on them, the excitation conditions of the studied structures are important factors affecting the type of the PL spectrum, in particular, the intensity, spectral position and width of the PL lines of the PCs. In the present paper, the influence of optical excitation conditions on the spectral and temporal characteristics of the PL of Ge(Si)-islands interstitial in two-dimensional PCs was investigated on the example of one of the narrowest PL lines observed in the spectra of the studied structures with PCs. It is shown that one of the main factors influencing the spectral position and shape of the PL lines, as well as the PL kinetics of photonic crystals with Ge(Si)-nano-islands, along with the structure of the PC modes, are the local heating of the investigated samples and the concentration of non-equilibrium charge carriers created by absorption of excitation radiation.

2. Experiment procedure

The initial structure with self-forming Ge(Si)-nanoislands was grown by molecular-beam epitaxy on a SOI substrate with a 2 μm thick buried oxide layer and a 70 nm thick Si top layer. The grown structure included a 50 nm thick Si buffer layer, an active area consisting of 5 layers of Ge(Si)-islands separated by 17 nm thick Si layers, and a 135 nm thick Si cover layer. The total thickness of the structure over the SiO_2 layer was 335 nm. The growth temperature of the islets was 600°C. The obtained structures exhibit the PL signal of Ge(Si)-islands in the spectral range of 1.3–1.6 μm [10,21]. The parameters and growth conditions of the structures were chosen based on the results of previous studies, which showed that under these conditions, the PL intensity of Ge(Si)-islands at room temperature is maximal [10].

PCs with hexagonal lattice of 50 × 50 μm with different periods and radii of holes were formed on the obtained structure by electron lithography and plasma chemical etching. The PC period ranged from 500 to 700 nm, and the ratio of hole radius to PC period was 0.2 or 0.25. The etching depth of the holes forming the PC in the structure considered in this paper was 250 nm. Details of the preparation of structures with PC, as well as their basic luminescent properties, in particular, the dependence of the PL spectra of Ge(Si)-islands in the PC on the PC parameters, are given in [17,18,20,21].

The spectro-kinetic PL dependences of structures with Ge(Si)-islands interstitial in PCs were studied by micro-PL spectroscopy with subnanosecond temporal resolution (Fig. 1). A picosecond laser system based on an Nd:YVO_4 laser („Solar LC“) with an emission wavelength of 532 nm, an $\sim$ 10 ps pulse duration, and a pulse repetition rate of 80 MHz was used for PL excitation. If necessary, the pulse repetition rate was reduced to 1 MHz using a laser pulse thinning device based on an electro-optical modulator. Focusing of the excitation radiation on the surface of the investigated structures, depending on the

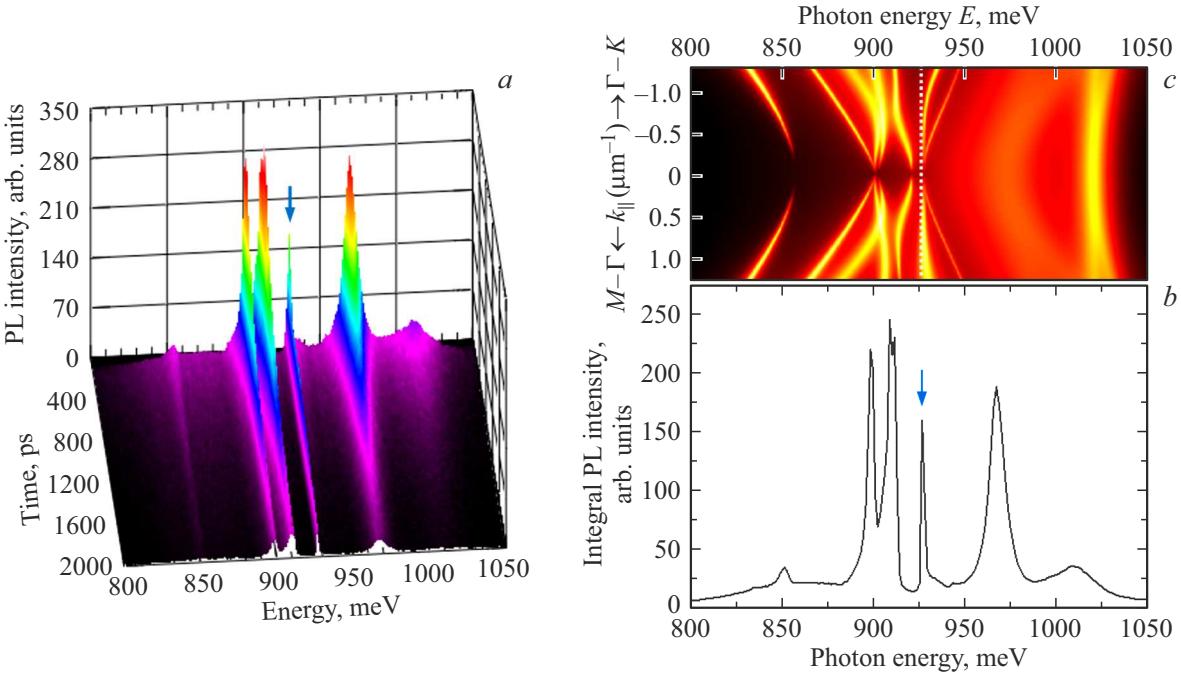

Figure 2. Spectro-kinetic dependence of PL (a) and time integral spectrum of PL (b) of photonic crystal „$a = 550$ nm, $r/a = 0.25$, $h = 250$ nm“ at $T = 295$ K, as well as calculated dispersion dependence of PC modes with these parameters (c). The narrowest line in the PL spectra (927.5 meV) in the plots (a) and (b) is indicated by an arrow, and the corresponding mode „E_2^{up}“ in the dispersion characteristic of the PC — dashed line.

required conditions of optical excitation, was carried out in two ways. In first case, both focusing of the laser beam and collection of the PC radiation were performed using a Mitutoyo Plan Apo NIR near-infrared lens with optical magnification 10× or 20×. The excitation spot size d was $\sim 5\,\mu$m (for lens 10×) or $\sim 2\,\mu$m (for lens 20×) (see insert to Fig. 1). In the second case, these lenses were used only to collect radiation from the studied structures, whereas the laser beam was focused using a long-focus lens ($F = 20$ cm) into a spot with a diameter of $d \sim 50\,\mu$m, which allowed relatively homogeneous excitation over the entire area of the studied PC ($50 \times 50\,\mu$m).

An Acton 2300i (Acton Research) lattice monochromator, a system for detection of individual photons based on a superconducting single-photon detector („Skontel“), and a TimeHarp 260 system (PicoQuant) for time-correlated counting of single photons were used to record the spectra and kinetic dependences of PL. The temporal resolution of the PL registration system was ~ 50 ps, and the spectral resolution was ~ 0.4 MeV. All PL measurements were performed at room temperature.

The dispersion dependence of the emissivity of the PC modes was calculated by the Fourier-modal method in the form of a scattering matrix [23].

3. Experimental results and discussion

In paper, the spectro-kinetic dependences of PL (PL intensity dependence on the photon energy and the delay time after the pump radiation pulse) for PCs with different parameters were investigated. In contrast to the PL spectrum of the initial structure with Ge(Si)-islands, which is a broad band in the range from 750 to 950 meV (1300 to 1650 nm) [10,21], the PL spectra of the PC are a series of lines of different widths corresponding to the PC modes, the intensity and spectral position of which are determined by the PC parameters [17,21]. Fig. 2, a shows the spectro-kinetic dependence of the PL intensity of PC with period $a = 550$ nm, ratio $r/a = 0.25$ and etching depth of PC holes $h = 250$ nm. It was found [22] that for these parameters, the narrowest PL line (width < 1 meV) is observed in the PL spectrum of PC, corresponding to the „BIC model“, labelled in [18] as „E_2^{up}“. Fig. 2, b shows the integral PL spectrum of this PC obtained by integrating the spectro-kinetic dependence of the PL intensity over the whole time interval considered. For comparison, Fig. 2, c shows the calculated dispersion dependence of the PC modes with these parameters at $T = 295$ K. The narrowest line in the above PL spectra (927.5 meV) is indicated by the arrow, and the corresponding mode „E_2^{up}“ in the dispersion characteristic of the PC is indicated by the white dashed line.

A detailed study of the influence of optical excitation conditions on the position and shape of the PL PC lines was carried out in this paper using the example of the indicated PL line. For this purpose, several variants of excitation of the studied structures were considered. As the first variant, let us consider the case of homogeneous excitation of the PC area with a pulse repetition rate of

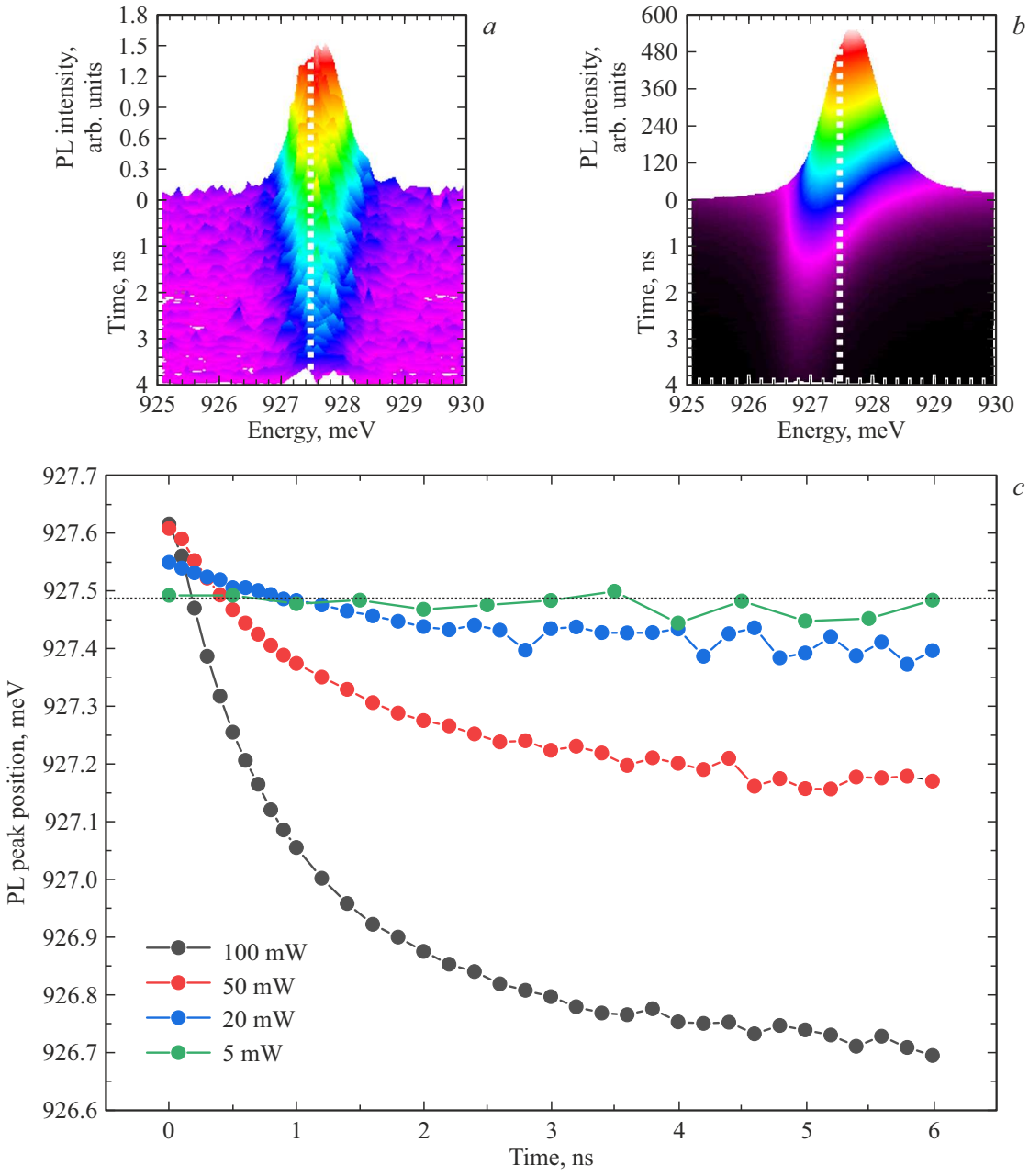

Figure 3. Spectro-kinetic dependences of the PL of PCs „$a = 550$ nm, $r/a = 0.25$, $h = 250$ nm“ in the vicinity of the line „E_2^{up}“, obtained by homogeneous PC excitation ($d = 50\,\mu$m) with a pulse repetition rate of 80 MHz and an average power of 5 mW (a) and 100 mW (b). c — time dependence of the spectral position of the „E_2^{up}“ line for different excitation power. The dotted lines show the „equilibrium“ position of the PL line (927.5 meV) obtained in the limit of small excitation power values.

80 MHz. In this case, a lens with a focal length of 20 cm was used to focus the laser beam, and the excitation spot diameter was $\sim 50\,\mu$m, which coincided with the size of the studied PCs. The use of a high pulse repetition rate (80 MHz) provides a high accumulation rate when measuring the time dependences of the PL signal with the help of the system of time-correlated counting of single photons; therefore, this mode of measurements is preferable for the study of PL with characteristic times < 10 ns (the repetition period corresponding to the frequency 80 MHz is 12.5 ns). In particular, this pulse repetition rate can be used to study the PL of PCs with Ge(Si)-islands at room temperature, since, as was shown earlier [20,21], in this case the characteristic times of the PL decline are from several hundred picoseconds to several nanoseconds (see Fig. 2, a).

Fig. 3 shows the spectro-kinetic dependences of PC PL „$a = 550$ nm, $r/a = 0.25$, $h = 250$ nm“ in the vicinity of the „E_2^{up}“ line obtained under conditions of homogeneous excitation over the PC area (excitation spot diameter $d = 50\,\mu$m) with a pulse repetition rate of 80 MHz and an

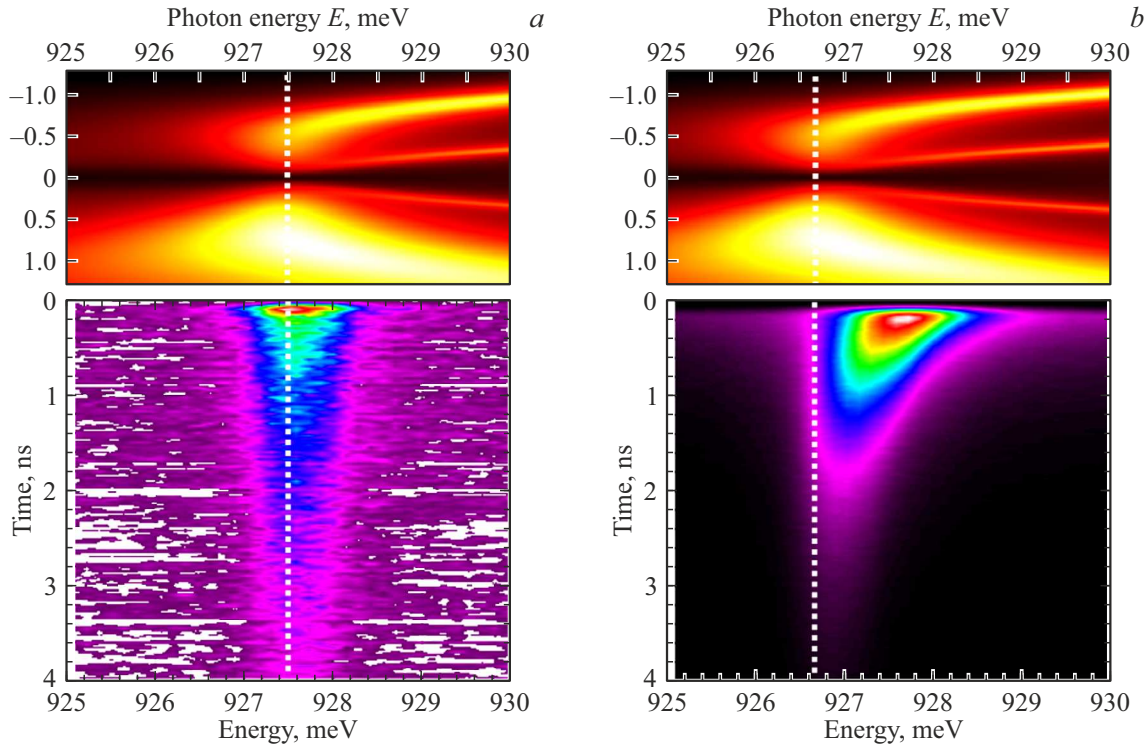

Figure 4. Comparison of the spectro-kinetic dependences of the PL line „E_2^{up}“ under homogeneous PC excitation with a repetition rate of 80 MHz and an average power of 5 mW (a) and 100 mW (b) with the calculated dispersion dependences of PC in the vicinity of this line at temperature values of 295 K (a) and 315 K (b). The dotted lines show the spectral position of the PL line at long delay times after the excitation pulse ($t = 6$ ns), which is 927.5 meV for $P = 5$ mW (a) and 926.7 meV for $P = 100$ mW (b).

average power of 5 mW (Fig. 3, a) and 100 mW (Fig. 3, b). As can be seen from the comparison of the obtained spectra, two opposite effects are observed with increasing excitation power. First, the increase in power leads to a noticeable broadening and shift of the PL line to the high energy area at the initial (in time) section of the spectro-kinetic dependence of the PL. At high excitation levels, a sharp dependence of the spectral position of the maximum of the PL line on the delay time after the end of the laser pulse is observed. This dependence at different average optical excitation power is presented in detail in Fig. 3, c. This effect seems to be caused by the dependence of the spectral position of the PL lines of photonic crystals on the concentration of non-equilibrium charge carriers, since under conditions of intense optical excitation, this value can make a significant contribution to both the real and imaginary parts of the refraction index and, as a consequence, lead to a change in the position of the PC lines.

The second effect observed at high excitation power is a change in the spectral position of the „tail“ of the spectro-kinetic dependence of the PL, i. e., the PL line position at large delay times after the excitation pulse (t). As can be seen from Fig. 3, c, at an average excitation power of 100 mW, the position of the PL line at $t = 6$ ns is 926.7 meV, i. e. by 0.8 meV less than the „equilibrium“ position of this PL line (927.5 meV) observed at low excitation levels (marked by the dashed line in Fig. 3, c). This shift is obviously not related to the influence of non-equilibrium charge carriers, since their concentration at large delay times is negligibly small, which is confirmed by the low level of the PL signal at this time interval (see Fig. 3, a and b). It is natural to assume that the observed red shift of the PL line is caused by local heating of the investigated structure in the area of the excitation spot, i. e., within the area of the considered PC. According to the calculations of the PC gap structure, an increase in temperature does lead to a shift of the PL lines of PC toward lower energies. In this case, as shown in Fig. 4, the value of the PL line shift equal to 0.8 meV corresponds to a change in the PC temperature from 295 to 315 K, i. e., a local heating of the PC by 20 K, which is a sufficiently plausible estimate.

Let us now consider the dependence of the shape and spectral position of the PL line on the repetition rate of laser pulses. As mentioned above, the use of a large pulse repetition rate (80 MHz) is optimal for the study of PL with characteristic decay times < 10 ns. At the same time, to study slower recombination processes (e. g., recombination of charge carriers in Ge(Si)-islands at low temperatures), it is necessary to increase the repetition period, i. e., to decrease the repetition rate of optical excitation pulses. In our measurement scheme, a laser pulse thinner is used for this purpose, allowing the pulse repetition rate to be varied from 1 MHz to units Hz. Fig. 5, a and b show

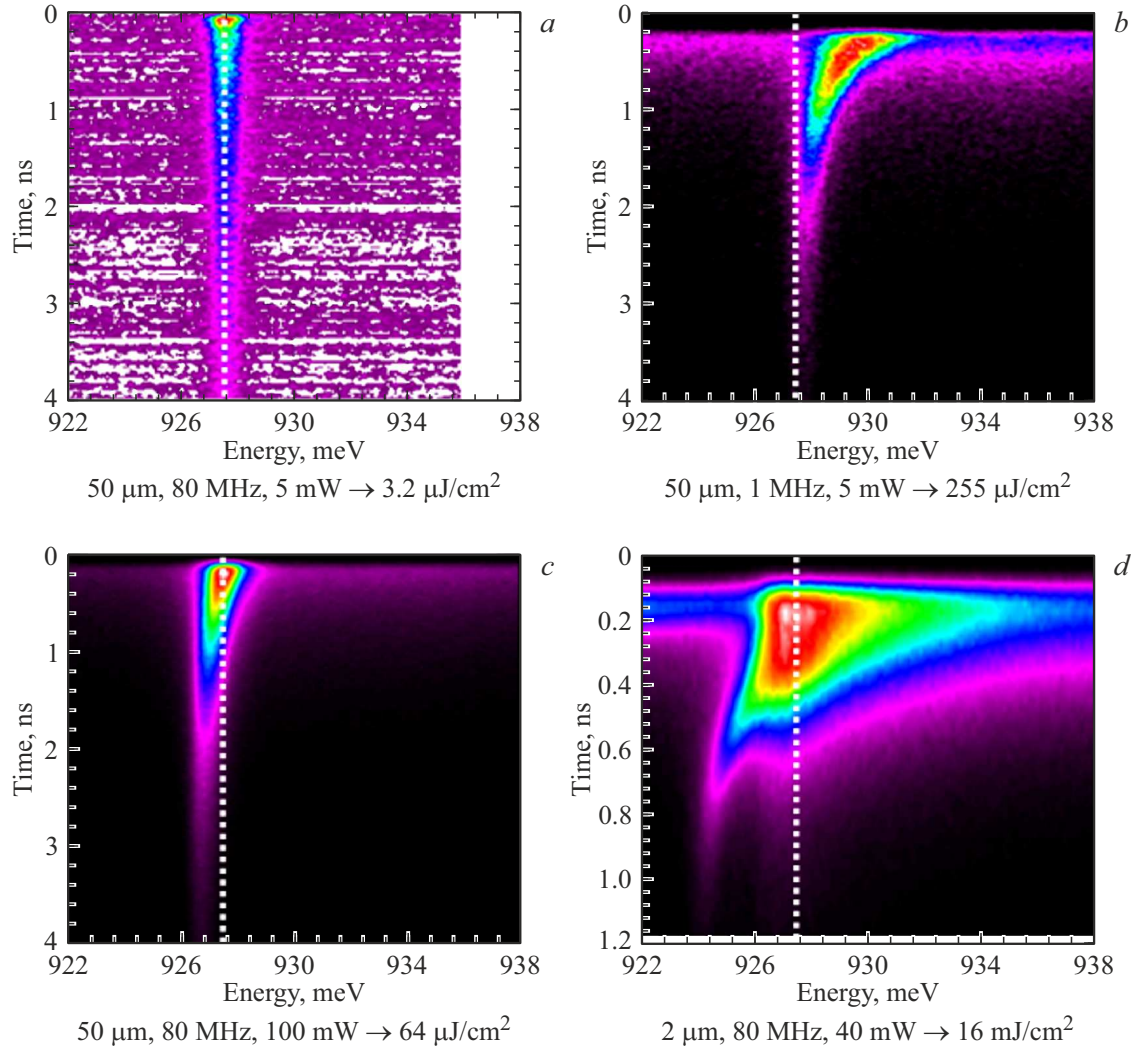

Figure 5. Spectro-kinetic PL dependences of the „E_2^{up}“ line obtained under different optical excitation conditions (indicated in the figure). The dotted lines show the „equilibrium“ position of the PL line (927.5 meV) observed at low levels of optical excitation.

the spectro-kinetic dependences of the PL line „E_2^{up}“ under homogeneous PC excitation with an average power of 5 mW and with different laser pulse repetition rates: 80 MHz (Fig. 5, *a*) and 1 MHz (Fig. 5, *b*). It should be noted that the mentioned variants of optical excitation are characterized by a significantly different optical excitation density due to the higher energy in the pump radiation pulse. At a repetition rate of 80 MHz, the optical pumping density was 3.2 μJ/cm^2 at an average excitation power of 5 mW and 64 μJ/cm^2 at a maximum power of 100 mW (Fig. 5, *c*), whereas at a repetition rate of 1 MHz at an average excitation power of 5 mW, this value was 255 μJ/cm^2. As a result, in the obtained spectro-kinetic dependence of the PL line „E_2^{up}“ at a repetition rate of 1 MHz, a much stronger broadening of the PL line in the area of high energies and time dependence of the position of the maximum of the PL line (Fig. 5, *b*) are observed, reflecting a significantly higher concentration of non-equilibrium charge carriers in the excitation area during absorption of pump radiation pulses. At the same time, the identical position of the PL line at long delay times after the excitation pulse (cf. Fig. 5, *a* and *b*), corresponding to the „equilibrium“ position of the PL line (927.5 meV) at low levels of optical pumping, indicates the absence of a noticeable local heating of the structure under these excitation conditions.

The measurements described above were carried out at the excitation spot diameter $\sim 50\,\mu$m, which allowed to realize almost homogeneous excitation over the area of the investigated PC. At the same time, in most studies carried out by micro-PL spectroscopy, focusing of the excitation radiation is carried out using the same lens with high optical magnification with which the radiation of the investigated microstructures (in our case — PC) is collected. The characteristic size of the focus spot ranges from 5 μm for a lens with 10x optical magnification to $\sim 1\,\mu$m when using lenses with higher optical magnification (20×, 50×). Under these excitation conditions, the inhomogeneity of excitation over the area of the investigated structures

becomes essential, which can lead to significant modification of the obtained spectro-kinetic dependences, especially at high levels of optical pumping.

Fig. 5, *d* shows the spectro-kinetic PL dependence of the „E_2^{up}“ line obtained at the maximum permissible average power of optical excitation (40 mW) with a pulse repetition rate of 80 MHz under conditions of focusing the laser beam into a spot of $\sim 2\,\mu$m size using the lens 20×. The energy density of the excitation radiation was 16 mJ/cm^2, i. e., more than 3 orders of magnitude higher than the minimum value (3.2 μJ/cm^2) obtained under homogeneous excitation of PCs considered in this paper. As can be seen from Fig. 5, *d*, at such extreme levels of optical excitation, the broadening of the considered PL mode line „E_2^{up}“ towards high energies reaches 15 meV. To quantitatively assess the contribution of non-equilibrium charge carriers to the high-frequency shift of the PL line of PCs under these excitation conditions, it is necessary to perform additional calculations of the gap structure of PCs, taking into account the high concentration of non-equilibrium charge carriers, including the possible occurrence of electron-hole plasma in the structures under consideration [24].

As can be seen from Fig. 5, *d*, at large delay times (t), the occurrence of two peaks with maxima of the PL intensity at photon energies 924.3 and ~ 927 meV is observed in the obtained spectro-kinetic dependence. The position of the first maximum (as follows from the comparison with calculations of the PC gap structure) corresponds to a change in the PC temperature by ~ 80 K and, apparently, corresponds to radiation from the PC area in the vicinity of the excitation spot, obviously characterized by strong local heating. The second (wider) line, the maximum of which is close to the position of the PL line of the „E_2^{up}“ mode under conditions of low excitation power, is apparently due to radiation from the rest of the PC surface, the heating of which appears to be insignificant.

4. Conclusion

Thus, the influence of optical excitation conditions on the spectral and temporal characteristics of the emission of two-dimensional PCs with Ge(Si)-nano-islands has been investigated. It is shown that the important factors influencing the spectral position and shape of the PL lines, as well as the PL kinetics of PCs, along with the structure of PC modes, are the local heating of the investigated samples and the concentration of non-equilibrium charge carriers created by the absorption of excitation radiation. At the same time, high concentration of charge carriers at high optical pumping density leads to broadening and shift of the PL lines of PCs to the area of higher energies at the initial time section of the spectro-kinetic dependence of the PL, whereas local heating of the samples under study, on the contrary, leads to a general shift of the PL spectrum of PCs towards lower energies. These effects should be taken into account when measuring the characteristic PL decay times of structures with PC at high levels of optical pumping, especially under conditions of strong focusing of the laser beam. It is shown that the use of optical excitation homogeneous over the area of the PC allows to significantly reduce the influence of the above effects on the measured time dependences of the PL intensity, as well as the experimentally determined width of the PL lines of the PC. The latter can be important, for example, for correct measurement of the goodness of PC modes, especially the so-called BIC („bound-state-in-continuum“) modes, characterized by high goodness of fit and, as a consequence, narrow lines in the spectra of PL PC.

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Conflict of interest

The authors declare that they have no conflict of interest.

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