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Photoluminescence from an active planar optical waveguide made of silicon nanocrystals: dominance of leaky substrate modes in dissipative structures

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Abstract

Samples with the structure of asymmetric planar waveguides are fabricated by implanting Si^+ ions with energy of 400 keV and doses from 3 to $6 \times 10^{17}\,cm^{-2}$ into synthetic silica slabs. Broad photoluminescence spectrum is observed when collecting photoluminescence (PL) signal in the direction perpendicular to the plane of waveguides while PL collected from the edge of the sample reveals narrow (FWHM of 10–20 nm) polarization-resolved transverse electric (TE) and transverse magnetic (TM) peaks. This peculiar observation is explained using a theoretical model, developed within the framework of wave optics. The TE and TM modes are identified as radiative leaky modes of the planar waveguide. Conditions under which the narrow modes can be seen in the edge PL spectra are described.

1. Introduction

Silicon is a material with an indirect band gap and thus optically excited electrons which had relaxed to the conduction band minimum cannot directly recombine by a photon emission with holes created at the valence band maxima. Assistance of an optical phonon is needed to fulfill the *k*-conservation rule. This fact reduces efficiency of photoluminescence by orders of magnitude compared to materials with a direct band gap. For Si-based optoelectronic devices, it is however, essential to have silicon-based light emitters. After the first observation of luminescence from porous silicon [1], confined systems became good candidates for light emitters. Recently, researchers try to find optical gain in samples containing silicon nanocrystals (Si-NCs) for construction of a silicon-based source of a coherent optical field [2].

Samples containing Si-NCs are fabricated mostly in a form of a thin film with nanocrystals deposited on a SiO_2 substrate [3,4] or in a form of multilayers of Si-NCs separated by SiO_2 layers [5]. A layer with Si-NCs has a higher refraction index than silica substrate and thus it forms a core of an active waveguide. Photoluminescence (PL) of the Si-NCs is then affected by confinement of an optical field inside the waveguiding layer.

In our previous publications, we reported on a peculiar observation of unexpected polarization-resolved

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(TE and TM) narrow modes in PL spectra detected in the direction parallel to the waveguiding layer [6,7]. Such modes were independently observed by other groups in different waveguiding samples [3]. Acurate theoretical model is, however, still lacking.

Here we present a model of an active planar waveguide based on wave optics. The model describes well presence of the modes as well as their spectral positions, widths and other characteristics. Influence of these modes on the measurements of optical gain (by the Variable Stripe Length (VSL) method [8]) is investigated.

2. Experimental

Our samples were prepared by Si^+ ion implantation with energy 400 keV into synthetic silica slabs (~1 mm thick) with doses 3, 4, 5 and $6\times10^{17}\,cm^{-2}$. Peak excess concentration of Si atoms was up to about $1.4\times10^{22}\,cm^{-3}$. Samples were annealed for 1 h in N_2 ambient at 1100 °C and 1 h in forming-gas (5% H_2 in N_2) at 500 °C. By the procedure, thin layers (approximately 1 μm thick) of Si-NCs below the surface of SiO_2 slabs were prepared. The presence of Si-NCs with diameter between 4 and 5 nm in the annealed layers was proven by Raman scattering. The high-quality samples with homogeneous implantation dose over the whole surface area (several cm^2) were produced. The samples provide waveguiding in a planar waveguide formed by the layer of Si-NCs. The profile of the refraction index can be calculated from measurements of near IR transmission spectra [9].

In the PL measurements, the sample was excited by a cw laser beam from a He–Cd laser (325 nm) without focusing; the excitation intensity was approximately $0.3\,W/cm^2$ and thus no nonlinear effects are expected to occur in the core of the waveguide. A microscope (numerical aperture 0.075) connected to an imaging spectrograph with a CCD camera was used for detection of the light emitted by Si-NCs. All measurements were performed at room temperature.

Typical PL spectra (measured for the samples with doses 4 and $5\times10^{17}\,cm^{-2}$) are plotted in Fig. 1. The dotted lines refer to the PL spectrum collected from the plane of the sample, i.e. in the direction perpendicular to the plane of the waveguide. These are typical broad PL spectra of Si-NCs and their shape is almost independent of the implantation dose. PL spectra emitted from the edge of the waveguide measured in the direction parallel to the waveguide plane reveal much different structure—see solid lines in the graphs in Fig. 1. Two narrow peaks, the TE and the TM-polarized modes, are resolved, whose spectral positions depend on the implantation dose. Fig. 1 illustrates clear red-shift of the mode doublet with increasing dose.

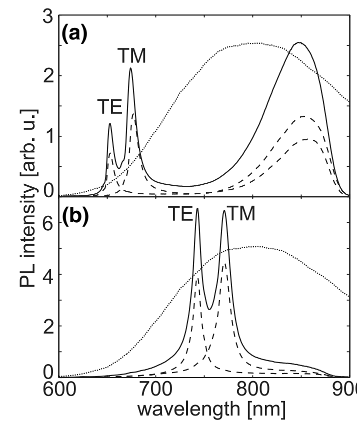

Fig. 1. Measured PL spectra from the edge of the samples with doses $4\times10^{17}\,cm^{-2}$ (a) and $5\times10^{17}\,cm^{-2}$ (b), numerical aperture was 0.075. The dashed lines show the TE and TM parts of the detected signal (detector was placed after an analyzer), the solid line is PL detected without the analyzer (both polarizations) and the dotted line shows the PL spectrum detected from the plane of the sample in the *x*-direction.

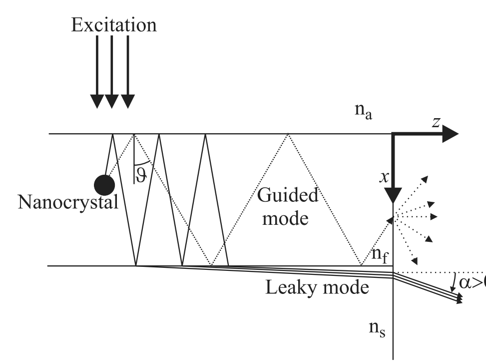

Fig. 2. Scheme of the propagation of rays emitted by an optically excited nanocrystal. The nanocrystal is embedded inside a layer with the refraction index n_f, this layer is deposited on a substrate with the refraction index n_s. Guided modes (dashed lines) cannot leave the core of the waveguide while the energy of the leaky modes (solid lines) is transferred into the substrate after several reflections on the boundary core/substrate.

In order to fully characterize the mode structure, we varied parameters of the experiment and we measured carefully the PL spectra. The parameters under variation were e.g. angle of observation, position of the excitation spot, numerical aperture of the detection system, implantation dose etc. [14]. These data were then used for comparison with the model presented below. We highlight here the most interesting result of those experiments: the peaks in the PL spectra were detected *only* at the angles of detection α (see Fig. 2) greater than 0.

3. Theory

Recent explanation of the occurrence of the modes in the PL spectra is based on the limit of single-mode guiding [10]. Khriachtchev et al. suppose that the second guided mode is mostly delocalized in the critical spectral area of the transition from the single to two-mode guiding. The more localized mode is, the higher losses are (if the core is lossy while the substrate transparent) and thus the mostly delocalized guided modes has the lowest losses and the highest contributions to the PL spectra. Since the model of weakly guiding waveguide was used, the model cannot describe the TE/TM mode splitting. Predictions of the spectral position of the modes is, however, in the spectral area of the detected modes in the experiments.

Even though the model presented in [10] gives reasonable arguments, no quantitative results were given and we believe that only the effect of localization of the guided modes cannot explain occurrence of the *narrow* peaks. In order to select important effects for the mode selection, we investigate carefully properties of light confinement by a waveguide.

Let's first assume a simple model of a three-layer asymmetric waveguide to show the basic properties of the active planar waveguides (see Fig. 2). A "core" layer with the refraction index n_f and thickness d is deposited on the substrate with the refraction index n_s. The system is surrounded by air (refraction index $n_a = 1$). The edge of the sample is perpendicular to the waveguiding layer.

A nanocrystal containing an excited electron-hole pair can be modeled by a dipole coupled to an electromagnetic field. The dipole is coupled only to the modes allowed by confinement of the electromagnetic field caused by the planar waveguiding structure. The allowed modes of the field are classified as the guided modes and radiative modes [11]. The guided modes are enclosed inside the core of the waveguide and they propagate in the z-direction without losses (their propagation constant is real). Contrary, the radiative modes loose their energy during propagation and thus the propagation constant has a nonzero imaginary part.

We use rays to demonstrate the propagation of light in Fig. 2. The light emitted by a nanocrystal proceeds towards one of the core/air or core/substrate boundary. It can be partly refracted into the air or substrate and is partly reflected back to the core. The light then propagates towards the second boundary and it again undergoes partial refraction and reflection. This scenario repeats until the ray inside the core looses all of its energy or it reaches the edge of the sample. Depending on the portion of the reflected light during reflection on the core boundaries, we distinguish three types of modes: (i) the guided modes are totally reflected during all reflections, (ii) the radiative modes are partly refracted into air during reflection on the core/air boundary and partly refracted into the substrate during reflection on the core/substrate boundary and (iii) the substrate radiative modes refract only into the substrate (they are totally reflected on the core/air boundary). If we introduce the propagation angle ϑ (see Fig. 2), we can write $\sin\vartheta_G > n_s/n_f$ for the guided modes, $\sin\vartheta_R < 1/n_f$ for the radiative modes and $1/n_f < \sin\vartheta_S < n_s/n_f$ for the substrate radiative modes. In addition to the simple ray model, light is absorbed and scattered by Si-NCs during propagation inside the core.

Ordinary theory of passive waveguides (light is coupled to a waveguide from outside) considers lossless media and thus sharp delta-like modes. As stated above, we assume some significant losses by absorption and scattering, therefore we expect relatively large linewidth of the allowed modes compared to the delta function. We then need to describe fully the spectral shape of the waveguide modes. We are going to use the theory presented in [12]. The intensity of light on the wavelength λ propagating at the angle ϑ from a dipole inside the waveguide is:

$$I(\vartheta,\lambda) = \frac{I_0(\lambda)}{|\,1 - R\exp[2ik(\lambda)d\cos\vartheta]\exp[-2\gamma(\lambda)d\cos\vartheta]|^2}, \quad (1)$$

where R stands for the product r_1r_2 of the reflection coefficients on the core/air and core/substrate boundaries, respectively, d is the thickness of the waveguide, $k = 2\pi n_f/\lambda$ is the wave vector of the light, γ is the coefficient of losses (scattering plus absorption) and $I_0(\lambda)$ is an intrinsic spectrum of the dipole radiation without confinement. It follows from the Fresnel formulas that the reflection coefficient R is small for small angles ϑ (radiative modes) and complex unity for guided modes. Guided modes then reveal sharp peaks while the radiative modes have a flat spectrum. The PL spectrum measured in the x-direction is then expected to represent the intrinsic PL spectrum of the Si-NCs.

Formula (1) predicts one important consequence concerning the guided modes: for any wavelength λ, there is an angle ϑ for which the denominator has a local minimum, i.e. in the region of the single mode guiding, there is one guided mode for all wavelengths. Provided that γ is independent of λ, the relative intensities of the modes would be equal. Detailed study of (1) under the above consideration revealed no narrow peaks in the overall spectrum of the guided PL even if the losses in the core were relatively high.

We have not discussed substrate radiative modes so far. These modes have the reflection coefficient R considerably greater than zero, i.e. for a fixed ϑ, the spectrum has minima and maxima. Near the critical angle for total reflection on the boundary core/substrate ($\sin\vartheta_c = n_s/n_c$), the value of the reflection coefficient R is close to unity. The spectrum of light emitted under this angle reveals narrow peaks. Because of different phase shifts during reflection on the core/air boundary, the TE and TM modes have slightly different spectral positions.

These substrate radiative modes reveal the mode structure because of a high reflection coefficient on the core/substrate boundary, but this reflection coefficient is still less than unity. It causes ''leaking'' of the energy into the substrate and thus those modes are called ''leaky modes''. The energy of the field transferred into the substrate leaves the core/substrate boundary as a plane wave and is no longer coupled to the field inside the core. This phenomenon is very similar e.g. to the radiation of a microcavity [13]: the confinement of the optical field selects allowed modes and a dipole transfers its energy only to those modes. Selecting the phase matching condition by tuning the angle of observation, PL modes shift in spectra and split. Once the light leaves the microcavity, it propagates freely and its energy is no more transferred back into the microcavity.

The basis of the presented model is the fact that the leaky modes are refracted into the substrate at angles near $\pi/2$. It means they propagate nearly parallel to the waveguide and leave the edge of a sample near the angle $\alpha = 0$, but always $\alpha > 0$ (due to the asymmetry of the waveguide—the mode cannot leak into the air layer above the waveguide). Since the PL detection is set to be in the z-direction, we expect to detect both guided and leaky modes together—the guided and the leaky modes are responsible for the continuum in the spectra and the distinct TE/TM mode structure, respectively.

4. Results of numerical calculations

For calculations, we use a realistic continuous profile of the refraction index of the waveguiding layers [9] found by fitting the IR transmission spectra with an asymmetric double-Gaussian curve added to a constant n_s.

We used the transfer matrix method for calculation of the intensity of the leaky modes and for calculation of the electromagnetic field distribution of the guided modes. Because of variation of refraction index, guided modes are not simply refracted at the edge of the waveguide from the core but a diffraction integral must be calculated. The intensity of the diffracted mode at the wavelength $\lambda = 713\,\text{nm}$ (spectral position of the TE mode) from the edge of the sample with the dose $5 \times 10^{17}\,\text{cm}^{-2}$ is plotted in Fig. 3. The curve has a maximum near the angle $\alpha = 0$, i.e. light is diffracted particularly in the z-direction. Energy collected by the detection system is determined by an integral over the collecting angle, therefore only a small portion of the energy of the guided modes can be registered by a detection system with small numerical aperture.

Result of a numerical simulation of the PL spectra from Fig. 1a are plotted in Fig. 4a. The narrow peaks originate from the leaky modes while the broad maximum at the right-hand side is due to the guided modes.

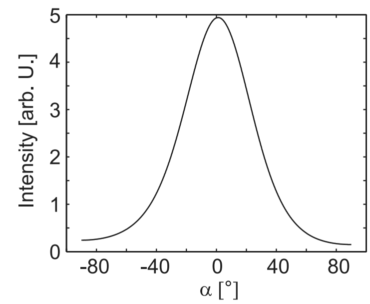

Fig. 3. Calculated dependence of the intensity of the diffracted guided mode with $\lambda = 713\,\text{nm}$ and $\vartheta = 55.3°$ on the angle of detection α. We used the parameters of the sample with the dose $5 \times 10^{17}\,\text{cm}^{-2}$ for the calculation.

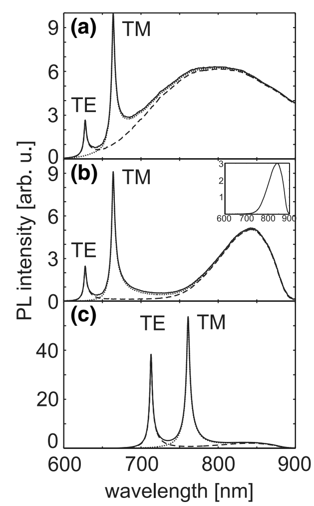

Fig. 4. Calculated PL spectra detected at $\alpha = 0$ for detector with a numerical aperture 0.075. (a) Leaky modes plus guided modes with the wavelength–independent coefficient of losses, dose $4 \times 10^{17}\,\text{cm}^{-2}$. (b and c) Leaky modes plus guided modes taking into account wavelength–dependent losses for the guided modes, doses 4 and $5 \times 10^{17}\,\text{cm}^{-2}$, respectively. We took the PL intensity measured from the edge of the sample with the dose $3 \times 10^{17}\,\text{cm}^{-2}$ (see inset) as a reference signal of the guided modes. The dashed lines stand for the TE mode, the dotted lines for the TM mode and the solid lines for the overall intensity.

The mode structure at the high-energy side of the spectra coincides well with the experimental data in Fig. 1a. However, the low-energy wings of the spectra (guided modes) differ—they are blue-shifted in the theory with respect to the experiment. We think that this feature is due to wavelength-dependent absorption, the onset of absorption by Si-NCs starting at approximately 850 nm.

To verify this hypothesis, we performed an experiment where we filtered detected light by a mask in a Fourier plane of a lens of the detector. By this filtering, only modes with $\alpha < 0$ passed into the detector so that the leaky modes were completely cut off. The spectrum is plotted in Fig. 5. This is the pure spectrum of the guided modes which is significantly different from the PL spectrum collected from the plane of sample.

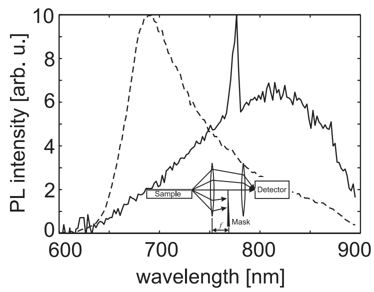

Fig. 5. Filtering of the leaky modes—the dashed line is the PL spectrum of the edge emission of the sample with the dose $4 \times 10^{17}\,\mathrm{cm}^{-2}$ without any filtering. The solid line is a filtered signal (see the inset) and represents the spectrum of the guided modes. The curves are normalized to unity. The numerical aperture of the detection system was 0.45 (significantly larger than for the PL spectra measurements in Fig. 1), the peak of the filtered signal is a doubled wavelength of an excitation laser.

The PL spectra from the Fig. 1a,b, respectively, were modeled assuming a waveguide-dependent coefficient of losses, see Fig. 4b,c, respectively. The spectrum of the guided modes after propagation through the waveguide was taken from the PL data measured on a sample with dose $3 \times 10^{17}\,\mathrm{cm}^{-2}$ (see inset of Fig. 4b) because this spectrum reveals no modes and our calculations have shown that it should be spectrum only of the guided modes. The experimental and theoretical spectra reveal a good agreement.

Validity of the presented theoretical model was further confirmed by numerous simulations of experiments performed on samples with different implantation doses, different angles of detection etc. [14].

5. Discussion

The ratio between the intensity of the leaky modes and the guided modes is driven by three main parameters:

1. Losses of the waveguide—the leaky modes propagate mostly in the lossless substrate. Contrary, intensity of the guided modes is exponentially decreasing when propagating towards the edge of the sample. Therefore the relative portion of the guided modes in the diffracted signal is reduced.
2. Spectral position of the leaky modes—as seen in Fig. 4b and c, the leaky modes are always more intense compared to the intensity of the guided modes at the same wavelength. But if the leaky modes are in the region of a weak intrinsic PL of the Si-NCs, they will have very low intensity compared to spectrum of the guided modes.
3. Numerical aperture of the detection system—the guided modes diffract at the edge of the sample while the leaky modes do not. Detecting in the z-direction (we collect the PL from an interval around the angle $\alpha = 0$), we collect the full energy of the leaky modes for selected angle ϑ and wavelength λ. In contrast, we detect only a small portion of the energy of the guided modes at the same wavelength due to small detection aperture. To detect as much energy of the guided modes as possible, we propose to use a big numerical aperture of the detecting system.

The combination of all three above mentioned effects results in a big contrast between the leaky and the guided modes in the modeled PL spectra (see for example Fig. 4c).

One of the most interesting points related to the leaky modes is whether or not light amplification can be achieved on them. On one hand, intensity of these modes can be high since they do not diffract at the edge of the sample. They propagate a limited path in the lossy waveguide core and thus they may escape from scattering/absorption on longer distances within the core. These may be favorable conditions for the occurrence of optical gain. Indeed, recently we have tentatively ascribed a weak net modal gain ($\approx 2\,\mathrm{cm}^{-1}$), observed in a sample implanted to a dose of $4 \times 10^{17}\,\mathrm{cm}^{-2}$ (Fig. 1a) under high density nanosecond excitation to the TM mode [15]. On the other hand, just because the leaky modes can be amplified on a relatively short path in the pumped core only, a significant optical amplification is probably hard to achieve. The leaky modes thus can play also a role of artifacts in the optical gain measurements using the VSL method [6]. It is likely that the occurrence of optical gain in the leaky modes can be affected by a considerable number of experimental parameters and as a such, it will deserve a particular analysis from case to case.

We propose experiments which enable to cut the leaky modes off when using the VSL method. The experimental setup is based on the asymmetry of the leaky mode emission. We propose a geometry where we cut all modes diffracted in the direction with $\alpha > 0$. The geometry is depicted in the inset of Fig. 5—a mask (a blade) is placed in the focal plane of a first lens or an objective (with a large numerical aperture) and the filtered light is then collected by a second lens (objective) onto a slit of a detection aperture. The intensity of the measured signal is reduced but all possible artifacts coming from the leaky modes are cut off. This approach can help to separate contributions from the guided and the leaky modes (see Fig. 5).

6. Conclusions

In this paper, we use theory of planar waveguides to explain a peculiar observation of narrow, polarization-resolved TE and TM peaks in PL spectra detected from the edge of a sample with the structure of a planar waveguide (this observation was firstly reported in [6]). The theoretical explanation of the PL measurements is based on the existence of leaky modes of the waveguide.

To compare theory and measurements quantitatively, we formulated a mathematical model and performed numerical calculations taking into account the real refraction index profile in the sample and wavelength-dependent absorption of the waveguide core. The results coincide well with the measured PL spectra—our model of the leaky modes is able to explain many different aspects of the experiments like TE/TM splitting, asymmetry in the spectra with respect to the angle of detection etc. We discussed how the leaky modes can manifest themselves in measurements of optical gain and proposed experiments with Fourier filtering in the focal plane of the detection system.

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