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J. Valenta, T. Ostatnický, I. Pelant, R. G. Elliman, J. Linnros, and B. Hönerlage

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Microcavity-like leaky mode emission from a planar optical waveguide made of luminescent silicon nanocrystals

J. Valenta and T. Ostatnický
Department of Chemical Physics & Optics, Faculty of Mathematics & Physics, Charles University, Ke Karlovu 3, CZ-121 16 Prague 2, Czech Republic

I. Pelant
Institute of Physics, Academy of Sciences of the Czech Republic, Cukrovarnicka 10, CZ-162 53 Prague 6, Czech Republic

R. G. Elliman
Electronic Materials Engineering Department, Research School of Physical Sciences and Engineering, Australian National University, Canberra ACT 0200, Australia

J. Linnros
Department of Microelectronics & Information Technology, Royal Institute of Technology, Electrum 229, S-164 40 Kista-Stockholm, Sweden

B. Hönerlage
IPCMS, Groupe d’Optique Nonlinéaire et d’Optoélectronique, UMR 7504 CNRS-ULP, 23, rue du Loess, 67037 Strasbourg Cedex, France

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The propagation of light emitted from silicon nanocrystals forming planar waveguides buried in SiO₂ is studied both experimentally and theoretically. Experiments reveal that photoluminescence spectra detected from the sample facet mainly contains narrow (10–20 nm full-with-at-half-maximum) polarization-resolved transverse electric and transverse magnetic modes instead of the usual broad nanocrystal emission band peaked at 700–800 nm. A theoretical model developed in the framework of wave optics identifies these modes as substrate modes propagating along the waveguide boundary (not the usual modes guided inside the nanocrystal plane due to its graded index profile). This peculiar observation is the consequence of the high losses in the nanocrystalline waveguide and may occur in other dissipative waveguide structures under conditions that are discussed. © 2004 American Institute of Physics. [DOI: 10.1063/1.1795984]

I. INTRODUCTION

Light-emitting silicon nanostructures have attracted considerable interest since the discovery of bright visible photoluminescence (PL) in porous silicon by Canham.¹ This interest has been further stimulated by recent reports of optical gain in an ensemble of silicon nanocrystals (Si-NCs).²–⁷ Most of the investigators looking for optical gain use samples in the form of a thin Si-NC layer (prepared either by the Si-ion implantation or by the plasma-enhanced chemical vapour deposition method) embedded in a transparent silica matrix, or Si/SiO₂ superlattices. Such structures work as asymmetric active planar optical waveguides, where the light is not coupled to the core from outside but originates from an ensemble of emitting Si-NCs inside the waveguide.

Here we report on experimental observations of unusual PL from planar waveguides made of luminescent Si-NCs in SiO₂ matrix. In particular, a significant narrowing of PL line-shape and splitting of the emission into spectrally separated transverse electric (TE) and transverse magnetic (TM) modes. Such behavior is very similar to III-V semiconductor-based microcavity PL TE-TM splitting observed in directions different from surface normal.⁸,⁹ We show that the narrow TE and TM modes, which the waveguide self-selects from a wide spontaneous emission spectrum, are not the usual guided modes, as assumed in our earlier report, but leaky substrate modes. They arise from constructive Fabry-Perot interference of luminescence rays leaving the active Si-NC layer to the substrate near the angle of total internal reflection. The existence of such modes is a general property of thin luminescent layers, irrespective of whether they are in the form of semiconductor microcavities,⁹,¹⁰ or simple organic/inorganic thin films.¹¹,¹² Hereafter, we deal with a general case of an asymmetric planar waveguide with a graded index profile where these modes are extremely well developed. We show, both experimentally and theoretically, that such experimental dominance of the leaky modes depends strongly upon a coincidence of several parameters, including the waveguide losses experienced by guided modes, the refractive index contrast of the core and cladding and the depth of core layer within the substrate.

II. SAMPLE PREPARATION AND EXPERIMENTAL SET-UPS

The samples used in this study were prepared by implanting 400 keV Si⁺ ions into 1 nm thick silica slab (Infra-sil) with optically polished surface and edges. Implantation...
doses of 3, 4, 5, and $6 \times 10^{17} \text{ Si cm}^{-2}$ (in four different regions of the slab) were chosen to produce different levels of refractive index contrast between the core and cladding/substrate layers. Peak excess Si concentrations were up to 26 at. % Si. Implanted samples were subsequently 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. The presence of Si-NCs in the annealed layers, with diameter between 4–5 nm, was confirmed by Raman scattering (not shown here).

The PL properties of samples were investigated using a continuous wave He-Cd laser (325 nm) as the excitation source (excitation intensity $\sim 0.3 \text{ W/cm}^2$) and a microscope connected to an imaging spectograph with a CCD camera for detection [Fig. 1(a)]. The detection numerical aperture (NA) was 0.075 (i.e., an angular resolution of about 8.6°). All measurements were performed at room temperature and all PL spectra were corrected for the system response. To achieve better angular resolution a second experimental arrangement was also employed in which the sample was fixed to the centre of a goniometer [Fig. 1(b)]. The PL emission was then collected by a silica optical fiber (core diameter 1 mm) rotated around the sample at a distance of 50 mm, giving an angular resolution slightly less than 1° (NA $\sim 0.01$). The output of the fibre was measured using the same detection system described above.

Typical images observed with the microscopic setup [Fig. 1(a)] are illustrated in Figs. 1(c) and 1(d). Here the diameter of the excitation spot, located at about 1 mm from the sample edge, is roughly 1 mm. One can easily recognize PL emission from the excited spot as a bright ellipsoid. However, there is also a second contribution emanating from the facet of the sample. This light is obviously guided in the implanted layer or close to it. The images in Figs. 1(c) and 1(d) were collected for sample inclination angles of $-15^\circ$ and $+15^\circ$, respectively, i.e., in a geometry for which the excited spot was observed either directly [Fig. 1(c)] or through the substrate [Fig. 1(d)]. The experimental arrangement shown in Fig. 1(a) enables the detection of the PL either from the excited spot or from the edge of sample by positioning the entrance slit of the spectrograph to different locations of the PL image.

### III. EXPERIMENTAL RESULTS AND DISCUSSION

Figure 2 (left column) presents PL spectra of four layers produced by implanting to different doses. The broad dotted curves correspond to PL emanating from the sample surface (perpendicularly to the Si-NC layer, i.e., conventional geometry) while the other curves represent PL collected from the facet of the sample (sample inclination $+2.5^\circ$). It can be seen that these two types of spectra differ considerably. The facet PL spectra are much narrower and contain (except the $3 \times 10^{17} \text{ cm}^{-2}$ sample) narrow TE and TM modes clearly re-

![Fig. 1](image1.png)  
**FIG. 1.** Sketch of the experimental geometry for the microPL setup (a) and for the goniometer setup (b). Lower panels show PL intensity images (area $2 \times 1.2$ mm$^2$) of the excited waveguide (using micro-PL setup) for sample inclination of $\alpha=+15^\circ$ (c) and $\alpha=+15^\circ$ (d). The elliptical spot is the excited region on the surface of the sample, and the narrow line is the PL emanating from the sample facet. The substrate is slightly illuminated with a white lamp giving rise to the lighter gray color in the bottom part of the images. The white arrows indicate the direction of excited beam.

![Fig. 2](image2.png)  
**FIG. 2.** Left column: Experimental PL spectra for layers implanted with doses 3, 4, 5, and $6 \times 10^{17} \text{ Si cm}^{-2}$ (from top down) detected in two different directions: The conventional normal incidence PL spectra (broad dotted line) and PL spectra emanating from the facet of the sample [black line—PL without polarizer, dashed line—with polarizer parallel to the layer (TE mode), short-dashed lines—polarizer perpendicular to the layer (TM mode)]. Right column: Calculated PL spectra from the sample facet ($\alpha =0^\circ$), taking into account the detection NA=0.075. The broad emission bands appearing in the samples $3 \times 10^{17} \text{ Si cm}^{-2}$ and $4 \times 10^{17} \text{ Si cm}^{-2}$ represent guided modes scattered into the detector collection angle.
solved with a linear polarization filter parallel (TE) or perpendicular (TM) to the layer edge. Such surprising PL spectra from a simple layer of Si-NCs have been reported for the first time only recently. Typical PL spectra from Si-NCs consist of a broad band centered at 700–800 nm with Gaussian-Lorentzian curves. The depth of the maximum is approximated either by nonsymmetric Gaussian or by Gaussian-Lorentzian curves. The angular dependence of the mode energy, i.e., the output PL intensity, is expressed as a function of the angle θ and wavelength λ. With the aid of equations for deriving the cavity enhancement factor, we get for the mode amplitude inside the waveguide

\[ A(\theta, \lambda) = A_0/(1 - t r_1 r_2), \]

where \( A_0 \) is an effective source strength and \( t < 1 \) is a transmission coefficient [in general \( t = t(\theta, \lambda) \)] during one roundtrip of the wave in the waveguide. \( r_1 \) and \( r_2 \) are the reflectivities of the waveguide boundaries that can be calculated (see, e.g., 16) by using a transfer-matrix method. The transfer matrix \( M \) is calculated as \( M = M_1 M_2 M_3 \cdots M_n \), where

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**FIG. 3.** Angular dependencies (sample \( 5 \times 10^{13} \text{ Si cm}^{-2} \)): Unpolarized PL spectra measured at various angles \( \alpha \) with respect to the normal to the output facet in the sample \( 5 \times 10^{12} \text{ Si cm}^{-2} \) using either the goniometer setup (a) or the micro-PL setup (b). The angular resolution is about 8.6° and 1°, respectively. Panel (c) represents model calculations of the angular dependence for aperture collection angle of 8.6°.

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**FIG. 4.** (a) Cross section of the Si-NC planar waveguide with a graded index profile \( n(z) \). Rays of both guided and substrate modes are indicated. (b) Refractive index profiles \( n(z) \) in the investigated samples. Lower two panels show calculated spectral positions of the substrate modes: As a function of the refractive index contrast \( \Delta n \) (c) and as a function of the maximum index contrast depth \( d \) in the sample \( 5 \times 10^{13} \text{ Si cm}^{-2} \) (d).
normal guided modes in an active planar optical waveguide. We have analyzed a waveguide made of Si-NCs but the effect is a general property of asymmetric thin films. The waveguide self-selects these modes from the broadband PL emission of the core. The spectral distribution and experimental manifestation of such modes is critically dependent upon several waveguide parameters, predominantly the refractive index difference between the core and cladding/substrate (determined by the implantation dose in the present study), the waveguide loss, the depth d of Si-NCs distribution beneath the surface and also upon the shape of n(z). The role of these modes in facilitating/hampering optical gain requires further analysis.\textsuperscript{11}

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\textsuperscript{15}The angle of incidence on the (sample/air) boundary is due to the graded index profile, greater than the critical angle \(\theta_0 = \arcsin(1/n(z=0))\) for all index profiles displayed in Fig. 4(b).
\textsuperscript{18}We stress that this is not an evanescent wave connected with the guided modes. Such waves cannot contribute to the far-field radiation. See E. Wolf and J. T. Foley, Opt. Lett. \textbf{23}, 16 (1998).
\textsuperscript{19}Note that the detection angle \(\alpha\) is linked to the angle \(\theta\) through \(\sin\alpha = n_{\text{substrate}}\sin(\pi/2 - \arcsin(n_{\text{substrate}}+\Delta n/2)\sin(\theta/n_{\text{substrate}}))\).