Active planar optical waveguides with silicon nanocrystals: Leaky modes under different ambient conditions

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We study both experimentally and theoretically the propagation of light emitted from silicon nanocrystals forming planar waveguides buried in SiO_2. Photoluminescence spectra detected from the sample facet show significant spectral narrowing—leaky modes—with respect to the spectra measured in standard photoluminescence configuration. The spectral position of the leaky modes responds strongly to a local change of refractive index (liquid drop) on the sample surface. Higher refractive index of the liquid induces higher redshift of the mode position. Experimental data agree with the previously proposed leaky mode model. © *2006 American Institute of Physics*.
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INTRODUCTION

Luminescent layers containing silicon nanocrystals are very promising materials for potential all-silicon optoelectronics. Strong effort is aimed nowadays towards successful demonstration of silicon-based laser. Several laboratories reported positive optical gain in systems with silicon nanocrystals.[1–8] Most of these samples are designed in a form of active planar waveguides. A significant narrowing of the photoluminescence (PL) emission spectrum measured in the direction of the waveguide plane (from the sample facet) is, among others, often used as an indication of the presence of stimulated emission in such waveguide structures. However, some recent works showed that in appropriate cases the significant PL spectrum narrowing does not result from stimulated emission, but from the development of a waveguide mode structure in the close vicinity of the waveguide cutoff frequency.[9–11] Such mode structure in PL spectra has so far been interpreted either as substrate leaky modes[12–14] or as a kind of delocalized guided modes near the cutoff frequency.[15–17] The question of which of the two models is valid deserves more detailed discussion.

In the present work, we study behavior of the nanocystalline-waveguide PL spectra at different ambient conditions and give further evidence in favor of the leaky mode model, developed previously by our group.[12,13] We change locally the refractive index (by dropping various liquids) on the sample surface above the excited luminescing spot and monitor subsequently spectral change of the PL. We compare the results with the theoretical spectra calculated using the leaky mode model. Excellent agreement of the experimental and theoretical data affirms the validity of the model.

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EXPERIMENT

The samples used in this study were prepared by implanting 400 keV Si^+ ions into 1 mm thick silica slab (Infrasil, refractive index $n_s = 1.455$) with optically polished surface and edges. Implant fluences of 3, 4, 5, and 6×10^{17} cm^{-2} were applied in four different regions of the slab. They produced different levels of refractive index contrast (with asymmetric graded index profiles[12]) 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 for 1 h in forming gas (5% H_2 in N_2) at 500 °C. Raman scattering confirmed the presence of Si nanocrystals in the annealed layers, with diameter between 3 and 6 nm.[18]

The PL properties of the samples were investigated using a continuous wave He–Cd laser (442 nm) as an excitation source. A silica optical cable collected the PL radiation. The output of the cable was connected to an $f = 20$ cm spectrograph equipped with a cooled charge-coupled device (CCD) camera. All measurements were performed at room temperature and all PL spectra were corrected for the system spectral response.

RESULTS AND DISCUSSION

Figure 1 recalls the peculiar waveguiding properties in our Si^+-ion implanted samples. The spectrum recorded in the standard 45° PL geometry [see thin solid line in Fig. 1(c) for the sample implanted to a fluence of 5×10^{17} cm^{-2}] consists of one broad peak (full width at half maximum of ~150 nm) centered at ~860 nm. However, the PL spectra detected from the cleaved facet of the sample—in the waveguide geometry—show, besides the broad peak due to ordinary guided modes, a fundamentally different feature: doublet of two narrow [full width at half maximum (FWHM) of ~20 nm] peaks. Here, the short-wavelength peak is TE po-

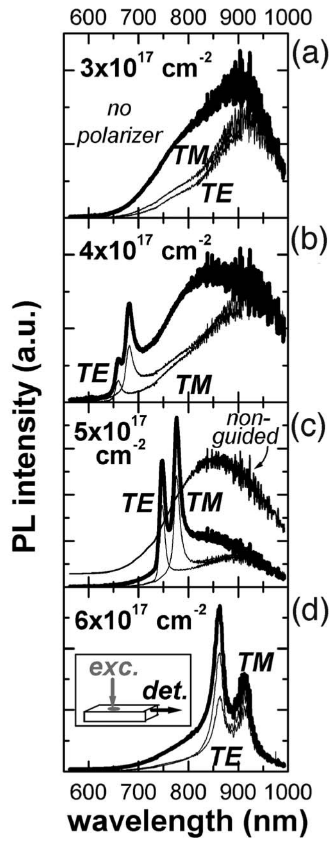

FIG. 1. Room-temperature PL spectra from the sample facet (in the waveguiding geometry schematically depicted in the inset) for layers implanted to a fluence of (a) $3\times10^{17}\ \mathrm{cm}^{-2}$, (b) $4\times10^{17}\ \mathrm{cm}^{-2}$, (c) $5\times10^{17}\ \mathrm{cm}^{-2}$, and (d) $6\times10^{17}\ \mathrm{cm}^{-2}$, compared to the conventional 45° PL geometry spectrum [curve "nonguided" in panel (c)]. Thick lines—PL without polarizer and thin lines—measurement with a polarizer: TE polarization (vector $\boldsymbol{E}$ parallel to the waveguiding layer) and TM polarization ($\boldsymbol{E}$ perpendicular to the layer).

larized (vector $\boldsymbol{E}$ parallel to the sample plane) while the peak on the long-wavelength side is TM polarized (vector $\boldsymbol{E}$ perpendicular to the sample plane). The position of this doublet shifts with increasing implant fluence (increasing refractive index contrast between substrate and the waveguiding layer) towards longer wavelengths.

We interpret the narrow modes in the edge PL spectra using the leaky mode model[12,13] schematically depicted in Fig. 2. The waveguide refractive index profile can be approximated by nonsymmetrical Gaussian or Gaussian-Lorentzian curves (similar to the implanted Si-ions distribution) with FWHM of about 0.3 μm.[12] The trajectory of relevant optical waves emitted by a chosen Si nanocrystal is shown. In case of ordinary guided modes, the optical wave undergoes total reflections both on the sample surface and at the interface waveguide core/substrate and the wave propagates inside the waveguide core. The leaky modes, on the contrary, are developing in a different way: optical wave, emitted by a silicon nanocrystal to the suitable direction (close to the boundary for the total reflection on the sample surface), undergoes the total reflection on the sample surface only. On the interface between the waveguide core and the substrate, where the refractive index contrast is lower than on the sample surface, the condition for the total reflection is not fulfilled. The light partially reflects and partially refracts at the angle very close to 90°; the refracted part then propagates outside the waveguide core (leaky or radiation mode) but almost parallel to it. The reflected part of light reflects again on the sample surface and interferes with the original refracted wave. The constructive interference arises only for a narrow range of wavelengths. Therefore, only narrow spectral range fulfilling the condition for the constructive interference is selected from the broad PL spectrum and these leaky modes manifest themselves in the PL spectra as very narrow peaks. Different spectral positions of the TE and TM polarized peaks can be then explained by different phase shifts for both polarizations, which are induced during the optical wave total reflection at the sample surface.

The question arises as whether ordinary waveguided light mode propagation within the implanted layer core can also occur in our samples. The answer is yes; this kind of emission can be noticed in samples implanted to fluences of 3×10^{17} and $4\times10^{17}\ \mathrm{cm}^{-2}$ as a wide band peaked at $\sim$850–900 nm [Figs. 1(a) and 1(b)]. However, these guided modes are strongly attenuated in samples implanted to higher total fluences due to waveguide losses. The exact nature of this attenuation is not known at present.[14]

From the point of view of the waveguide optics, we expect the leaky modes to be spectrally situated at slightly lower frequencies than the cutoff frequencies of the waveguide. This is the main observable difference between our leaky mode theory and the approach based on delocalized ordinary waveguide modes as proposed by Khriachtchev *et al.*,[15–17] where these modes approach the cutoff frequency from the higher frequency side. Because values of theoretical cutoff frequencies are not easy to calculate, in particular, for graded index profile, neither is easy to distinguish between the two above models on the basis of spectral PL measuremens themselves. However, we have recently proposed and realized a simple experimental approach of how to do it, which takes advantage of local change of refractive index on the sample surface.[14] In what follows we apply this method in a modified form to investigate further the properties of the TE/TM doublets.

Figure 2(a) depicts the principle of our experiment, based on dropping various liquids onto the excited spot on the sample. By dropping a selected liquid, we change locally the refractive index of the surrounding media on the sample surface (air, formerly). The optical conditions for developing leaky modes will thus be changed on the sample surface. The optical wave travels different distances and undergoes different phase shifts during total reflection. Therefore, also the conditions for constructive interference forming the leaky modes change, which should manifest directly in the PL spectra as a shift of the observed modes. Indeed, the left column in Fig. 2(b) shows the change of the PL spectra in our quartet of the samples upon dropping ethanol onto the sample surface. In all cases, the observed narrow modes undergo a significant redshift.

The right column of Fig. 2(b) demonstrates clearly that the above-mentioned leaky mode model is able to describe the observed redshift of the modes with high fidelity. This column presents the results of theoretical calculations of the

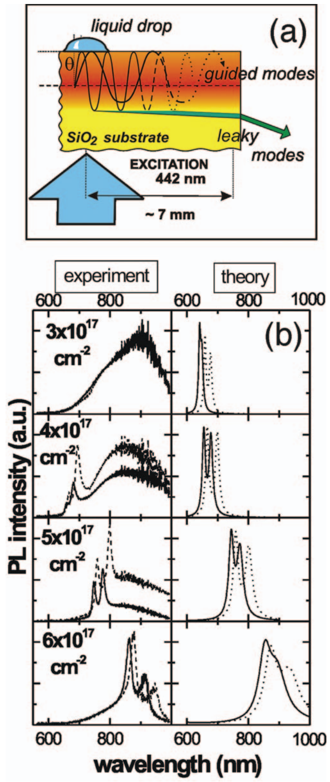

FIG. 2. (Color) (a) Schematic cross section of the asymmetric planar waveguide showing propagation of the guided modes as well as formation of the substrate leaky modes. Surface refractive index change (induced by a liquid drop placed directly above the excited region) influences the development of the leaky modes. (b) Comparison of the PL spectra in the waveguiding geometry for the samples in ambient atmosphere (full lines) and upon dropping ethanol (refractive index $n=1.361$, dotted lines) on the sample surface. Implant fluences are indicated for each sample. Left column: experimental data and right column: theory of leaky modes.

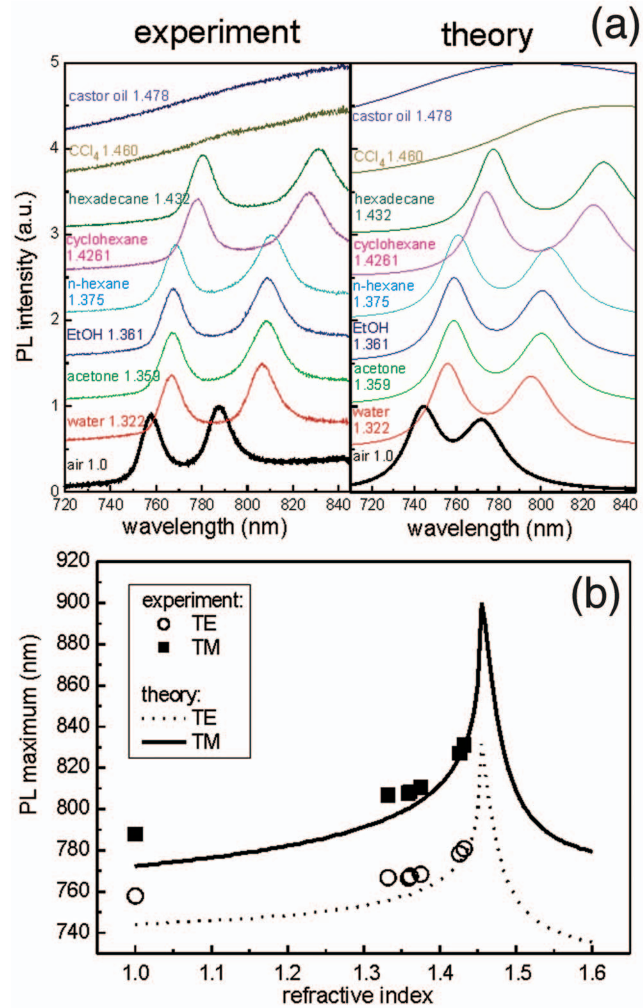

FIG. 3. (Color) (a) PL spectra in the waveguiding geometry showing leaky modes in the layer implanted to a fluence of 5×10^{17} cm^{-2}. Drops of various liquids above the excited spot lead to a redshift of both TE and TM modes. Spectra corresponding to various liquids have been vertically shifted. (b) PL peak position as a function of the refractive index of the liquid. Symbols: experimental data and lines: theory. For liquid refractive index higher than refractive index of the silica substrate ($n_s=1.455$) the theory predicts both disappearance of the distinct doublet structure and a back shift of the broad emission band to shorter wavelengths [see also the right panel in (a)].

leaky mode model developed in the framework of wave optics. In calculating these curves the above mentioned graded index profile of each sample, as determined by fitting interference-modulated optical transmission spectra of the implanted layers, was taken into account, together with refractive index values of applied liquids. Neither spectral profiles nor spectral positions of the substrate leaky modes can be calculated analytically. Numerical calculations were performed using the formula for cavity enhancement factor[19] (for more details, see Refs. 12 and 13) for the whole set of the samples. Taking in account that the theoretical model calculates only the leaky mode part of the PL spectra but not the ordinary, spectrally broad guided modes, both experimental data and the model correspond very well, which provides strong support for the validity of the model.

In order to further support the model, we investigate both experimentally and theoretically the effect of different liquids (different refractive indices) dropped onto the sample implanted to a fluence of 5×10^{17} cm^{-2}. The results are drawn in Fig. 3(a) and again, the measured and the simulated data agree very well. With increasing refractive index of the liquid, we initially observe increasing redshift of the modes. At some point, however, the "doublet" mode structure disappears and a broad PL spectrum can be seen. Actually, this happens when the refractive index of the liquid reaches the refractive index of the sample substrate ($n_s=1.455$), i.e., the point where the waveguide loses its asymmetry, total reflection on the upper boundary is canceled, and the condition for developing narrow TE/TM resolved leaky modes is not fulfilled anymore.

Figure 3(b) plots the PL peak position versus the refractive index of the applied liquids. Further refractive index increase above n_s still keeps the broad spectrum. Theoretically calculated shift goes, somewhat surprisingly, back to shorter wavelengths. However, this can be intuitively understood, since with further increasing refractive index of the liquid above the refractive index of the sample substrate, the role of the substrate and of the capping medium will interchange and (another type of) leaky modes should appear again. Such a back shift is, however, difficult to trace experimentally because of the large spectral bandwidth and possible admixture of normal incidence PL emission.

In calculating the theoretical curves in Figs. 3(a) and

3(b) we considered the liquid droplet thickness infinite, since its real thickness ($\sim$1 mm) is much larger than the waveguide core thickness as determined by the refractive index profile (FWHM of $\sim$0.3 μm).

CONCLUSIONS

In conclusion, by comparing the experimental and theoretical PL spectra under different ambient conditions, we further verified the validity of the leaky mode PL model in the samples containing silicon nanocrystals embedded in a SiO_2 matrix. This phenomenon can find practical applications, for example, as an optical sensor of the refractive index of the media surrounding the sample.

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[1] L. Pavesi, L. Dal Negro, C. Mazzoleni, G. Franzo, and F. Priolo, Nature (London) **408**, 440 (2000).
[2] L. Dal Negro *et al.*, Physica E (Amsterdam) **16**, 297 (2003).
[3] L. Dal Negro, M. Cazzanelli, L. Pavesi, D. Pacifici, G. Franzo, F. Priolo, and F. Iacona, Appl. Phys. Lett. **82**, 4636 (2003).
[4] L. Khriachtchev, M. Räsänen, S. Novikov, and J. Sinkkonen, Appl. Phys. Lett. **79**, 1249 (2001).
[5] J. Ruan, P. M. Fauchet, L. Dal Negro, M. Cazzanelli, and L. Pavesi, Appl. Phys. Lett. **83**, 5479 (2003).
[6] K. Luterová, K. Dohnalová, V. Švrček, I. Pelant, J.-P. Likforman, O. Crégut, P. Gilliot, and B. Hönerlage, Appl. Phys. Lett. **84**, 3280 (2004).
[7] M. H. Nayfeh, N. Barry, J. Therrien, O. Aksakir, E. Gratton, and G. Belomoin, Appl. Phys. Lett. **78**, 1131 (2001).
[8] K. Luterová, I. Pelant, I. Mikulskas, R. Tomasiunas, G. Muller, J.-J. Grob, J.-L. Rehspringer, and B. Hönerlage, J. Appl. Phys. **91**, 2896 (2002).
[9] J. Valenta, I. Pelant, and J. Linnros, Appl. Phys. Lett. **81**, 1396 (2002).
[10] K. Luterová, D. Navarro, M. Cazzanelli, T. Ostatnický, J. Valenta, S. Cheylan, I. Pelant, and L. Pavesi, Phys. Status Solidi C **2**, 3429 (2005).
[11] K. Luterová *et al.*, Opt. Mater. (Amsterdam, Neth.) **27**, 750 (2005).
[12] J. Valenta, T. Ostatnický, I. Pelant, R. G. Elliman, J. Linnros, and B. Hönerlage, J. Appl. Phys. **96**, 5222 (2004).
[13] T. Ostatnický, J. Valenta, I. Pelant, K. Luterová, R. G. Elliman, S. Cheylan, and B. Hönerlage, Opt. Mater. (Amsterdam, Neth.) **27**, 781 (2005).
[14] I. Pelant, T. Ostatnický, J. Valenta, K. Luterová, E. Skopalová, T. Mates, and R. G. Elliman, Appl. Phys. B: Lasers Opt. **83**, 87 (2006).
[15] L. Khriachtchev, M. Räsänen, and S. Novikov, Appl. Phys. Lett. **83**, 3018 (2003).
[16] L. Khriachtchev, M. Räsänen, S. Novikov, and J. Lahtinen, J. Appl. Phys. **95**, 7592 (2004).
[17] L. Khriachtchev, M. Räsänen, and S. Novikov, Appl. Phys. Lett. **86**, 141911 (2005).
[18] G. Viera, S. Huet, and L. Boufendi, J. Appl. Phys. **90**, 4175 (2001).
[19] R. Baets, P. Bienstman, and R. Bockstaele, in *Confined Photon Systems*, Lecture Notes in Physics Vol. 531, edited by H. Benisty, J.-M. Gérard, R. Houdré, J. Rarity, and C. Weisbuch (Springer, Berlin, 1999), p. 38.