

COMMENTS**Comment on “Enhancement of optical gain of semiconductors embedded in three-dimensional photonic crystals” [Appl. Phys. Lett. 71, 1616 (1997)]**S. G. Romanov^{a)}*Department of Electronics and Electrical Engineering, University of Glasgow, Glasgow G12 8QQ,**United Kingdom**Ioffe Physical Technical Institute, 194021, St. Petersburg, Russia*

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The original letter¹ addresses the impact of the photonic band gap (PBG) environment on the photoluminescence (PL) spectrum. PL of similar materials has been previously examined, particularly, by means of changing the excitation power,²⁻⁴ however the quantitative analysis was undertaken for the first time.

The transmission spectrum of CdS opal shows the deep minimum at 600 nm [Fig. 1(a)] for the light propagating normal to (111) plane of opal package. In accord with other reports for well-structured opals^{5,6} this minimum was interpreted as the stop band. The developing of the emission maximum near the stop band (*B* band at 570 nm) under the increasing power of excitation [Figs. 1(b), (c), and 3] was argued as the amplification of emission due to the PBG effect.¹

Opal impregnated with a semiconductor is known as the sort of templated nanocomposite, where the three-dimensional (3D) lattice of nanoparticles is formed by infilling structural voids of a crystalline insulator with “guest” material. PL spectrum of such composites consists of two contributions—from the infill itself and from the template. In the case of SiO₂ templates the latter is the PL band of oxygen defects. Usually, this band appears up to ten times enhanced after infilling template with guest material due to the effective energy exchange between guest nanoparticles and template defects.⁷ Recently, this mechanism was employed to understand the PL spectra of infilled zeolites, asbestos, and opals.⁸⁻¹⁰ For example, the enhancement of defect-related PL (bands at 563 and 730 nm, half-width 50–80 nm) was observed after the deposition of InP in different silica templates, structure of which does not exhibit Bragg resonance in visible light.¹⁰ Moreover, we emphasized the five times faster increase of the “green” PL band of InP asbestos (lattice period ~ 50 nm) upon the increasing pumping as compared with PL of bare asbestos.¹¹

Generally, the in-void chemical synthesis of any sort results in the nucleation of nanoparticles at defects of a template. Similarly, an interaction of reagents with defects of

template has occurred, when opal was treated with high-temperature vapors of Cd and S. This correlation permits the energy traffic from CdS nanoparticles to defects of opal, i.e., the indirect pumping of the PL at *B* band. CdS particles in the glass cannot be used as the standard for interpretation of PL of CdS opal, because the chemical routine employed for their preparation is principally different. Thus the PL spectrum of CdS opal can be thought as the superposition of band-edge recombination emission in CdS and the emission of opal defects, moreover the latter grows faster upon the pumping power. The weakness of the defect-related PL in the bare opal correlates with the above model.

Let me consider to the anisotropy of the PL spectrum. It is well documented that the suppression of the spontaneous emission at the stop band of incomplete photonic crystal appears as the dip in its PL spectrum.^{2,4,6,12} The anisotropy of PL spectrum reveals the probability for emitted photon to escape from the photonic crystal in a given direction. All crystallographic planes contribute to the probability pattern simultaneously, but with the weight factor that is proportional to the density of scatterers. That is why the angle of the emission collection relatively to the densest packed (111) plane is the most relevant parameter.⁶

Accordingly to Ref. 1, the PL was excited through the (111) base plane of the sample and the emission was collected from unidentified cleaved edge. Correspondingly, the PL spectrum should be determined by the angle θ with another (111) plane, which is nearly parallel to the cleaved side. Any direction in fcc crystal can be described by the polar angle φ for rotation around the [111] axis and the angle θ with this axis. The emission normal to (111) plane sees the lowest energy position of the stop band at 600 nm [Fig. 1(a)]. Rotating the strip by $\Delta\varphi=30^\circ$ at the illuminated plane is equivalent to changing of θ angle with regard to plane of collection. According to the authors' assumption the stop band is blue shifted, when $\Delta\varphi=30^\circ$ applies (along $\langle 110 \rangle$ direction). Correspondingly, if the *B* band is the band of amplified spontaneous emission, it should exhibit the same shift, but it does not. This behavior contrasts the correlation of the shift of the PL dip with the angular dependence of the stop band, that was clearly demonstrated for the bare opal,

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CdS opal, ZnS opal, etc.^{4,6,12} Therefore, the reasonable scenario is that φ rotation of the strip results in the overlap of the stop band in $\langle 110 \rangle$ direction with B band and, obviously, in the inhibition of spontaneous emission at the stop band. No suppression of the B band (at 570 nm) takes place in $\langle 211 \rangle$ direction because the stop band is centered at 600 nm (Fig. 3). In other words, the claim for three-time amplification of emission at the stop-band edge should be replaced with the declaration of three-time inhibition of emission within the stop band. The absence of the shift of B band in disagreement with the stop-band shift¹ assumes the independence of the B band upon the Bragg resonance. This conclusion agrees with the first part of Comment. Thus, the described experiment is insufficient to make the choice in favor of PBG relation of the B band.

¹Yu. A. Vlasov, K. Literova, I. Pelant, B. Honerlavage, and V. N. Astratov, Appl. Phys. Lett. **71**, 1616 (1997).

²J. Martorell and N. M. Lawandy, Opt. Commun. **78**, 169 (1990).

³S. G. Romanov, A. V. Fokin, V. V. Tretiakov, V. Y. Butko, V. I. Alperovich, N. P. Johnson, and C. M. Sotomayor Torres, J. Cryst. Growth **159**, 857 (1996).

⁴V. N. Bogomolov, S. V. Gaponenko, I. N. Gemanenko, A. M. Kapitonov,

E. P. Petrov, N. V. Gaponenko, A. V. Prokofiev, and S. M. Samoilovich, Phys. Rev. E **55**, 7619 (1997).

⁵R. Mayoral, J. Requena, J. S. Moya, C. Lopez, A. Cintas, H. Miguez, F. Merseguer, L. Vazquez, M. Horlado, and A. Blanco, Adv. Mater. **9**, 257 (1997).

⁶S. G. Romanov, A. V. Fokin, V. I. Alperovich, N. P. Johnson, and R. M. De La Rue, Phys. Status Solidi **163**, 169 (1997); S. G. Romanov, A. V. Fokin, N. P. Johnson, and R. M. De La Rue, Proceedings of the 197 Meeting of Electrochemical Society, edited by C. Ronda (unpublished).

⁷Yu. A. Alekseev, V. N. Bogomolov, T. B. Zhukova, V. P. Petranovskii, and S. V. Kholodkevich, Sov. Phys. Solid State **24**, 1384 (1982).

⁸S. G. Romanov, A. V. Fokin, V. Y. Butko, V. V. Tretiakov, S. M. Samoilovich, and C. M. Sotomayor Torres, Phys. Solid State **38**, 1825 (1996).

⁹S. G. Romanov, V. Y. Butko, H. M. Yates, M. E. Pemble, and C. M. Sotomayor Torres, J. Appl. Phys. **82**, 380 (1997).

¹⁰S. G. Romanov, H. M. Yates, M. E. Pemble, M. W. Anderson, J. R. Agger, V. Y. Butko, A. Y. Kumzerov, and C. M. Sotomayor Torres, Phys. Solid State **39**, 641 (1997).

¹¹S. G. Romanov and C. M. Sotomayor Torres, in *Nanoscale Science and Technology*, edited by N. Garcia, M. Nieto-Vesperinas, and H. Rohrer (Kluwer Academic, The Netherlands, 1998), pp. 255–270.

¹²S. G. Romanov, A. V. Fokin, and R. M. De La Rue, Appl. Phys. Lett. (to be published); S. G. Romanov, A. V. Fokin, and R. M. De La Rue, J. Porous Mater. (to be published).