# Strong modification of light emission from a dye monolayer via Bloch surface waves

Marco Liscidini,[1,*] Matteo Galli,[1] Molu Shi,[2] Giacomo Dacarro,[1] Maddalena Patrini,[1] Daniele Bajoni,[3] and J. E. Sipe[1,2,3]

[1]*Consorzio Nazionale Interuniversitario per le Scienze Fisiche della Materia and Department of Physics "A. Volta," University of Pavia, Via Bassi 6, I-27100 Pavia, Italy*
[2]*Department of Physics, University of Toronto, 60 St. George Street, Ontario M5S 1A7, Canada*
[3]*Department of Electronics, University of Pavia, via Ferrata 1, I-27100 Pavia, Italy*
**Corresponding author: marco.liscidini@gmail.com*

Received March 31, 2009; revised June 16, 2009; accepted July 3, 2009; posted July 9, 2009 (Doc. ID 109446); published July 24, 2009

We observe a strong modification of the emission properties of a rhodamine monolayer bonded to the surface of a 1D $Si_{1-x}N_x$:H PhC. The photoluminescence signal observed at 670 nm is enhanced by more than 1 order of magnitude at the angle where the emitters are evanescently coupled to a Bloch surface wave supported by the structure. These results may be important for the design of efficient light-emitting devices and of optical sensors. © 2009 Optical Society of America

*OCIS codes:* 230.5298, 300.2140, 240.6690.

Control of light emission from molecules at an interface is central to the design of a number of optical devices, including efficient light emitters and biosensors based on fluorescence [1,2]. Studies are often focused on metallic surfaces, where strong enhancement of the light–matter interaction is obtained by exploiting resonant coupling with surface plasmons (SPs) [3,4].

Another strategy for such control and enhancement is based on PhC (PhC) structures, where a periodic modulation of the dielectric function on a scale comparable to the wavelength of interest allows for the control of light propagation, including even confinement below the diffraction limit [5]. The possibility of exploiting the photonic bandgap (PBG) to inhibit spontaneous emission is at the center of pioneering work on PhC by Yablonovitch [6]. The suppression or enhancement of spontaneous emission in PhC structures, and the redistribution of the emitted light, are still the subjects of a host of current investigations [7–10].

Bloch surface waves (BSWs) are propagation modes that exist at the surface of a PhC [11]. These modes have a frequency within a PBG, so light cannot propagate through the periodic structure, and near-interface localization of the field is achieved owing to total internal reflection (TIR) at the surface of the bounding homogeneous medium. In many aspects, BSWs can be thought of as dielectric analogues of SPs, with the PhC playing the role played by the metal in confining the field of an SP near the interface [12]. Although BSWs have been known since the late 1970s, interest in the use of these surface waves is now growing [13–15], particularly for sensing and biosensing [16–21].

Exploiting the analogy between BSWs and SPs, in this paper we study the emission properties of a dye monolayer bounded to the surface of a one-dimensional PhC supporting a BSW.

We consider a periodic multilayer made of a-$Si_{1-x}N_x$:H, grown by plasma-enhanced chemical vapor deposition on a 7059 Corning glass substrate [22]. The first layer is 32 nm of a-$Si_{0.45}N_{0.55}$:H, followed by seven periods of a unit cell composed of 145 nm of a-$Si_3N_4$:H and 135 nm of a-$Si_{0.45}N_{0.55}$:H. The structure is designed to have a TE-polarized BSW in a range around 1.85 eV (670 nm) if the cladding is air. This falls within the emission range of rhodamine, and the multilayer is almost transparent at such frequencies. In Fig. 1 we plot the BSW and photonic gap frequencies for TE-polarized light as a function of the in-plane wave-vector component $k_x$ (see inset in Fig. 2). Since for a BSW the electromagnetic field is confined by TIR from the air side and by the PBG in the multilayer, the dispersion curve is below the air light line and within the PBG [12].

To study the interaction between an emitter and the BSW, we chose rhodamine as the dye molecule, which is characterized by a strong emission band centered at 2 eV (600 nm). We bonded a rhodamine monolayer to the PhC surface and to a reference

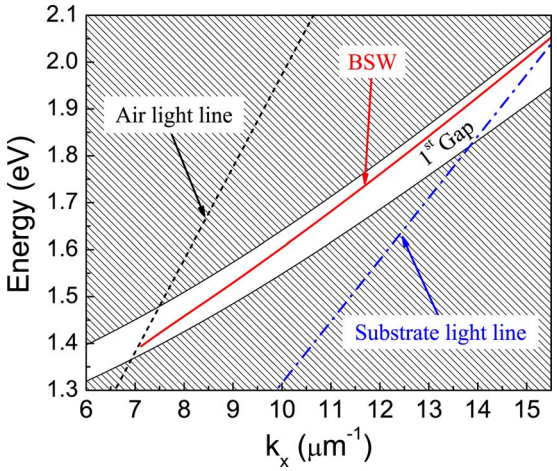


Fig. 1. (Color online) TE-polarized Bloch surface wave dispersion (solid red) for the a-$Si_{1-x}N_x$:H multilayer. Light lines for air (dashed black) and Corning 7059 ($n=1.55$) (dashed-dotted blue) are shown.

 

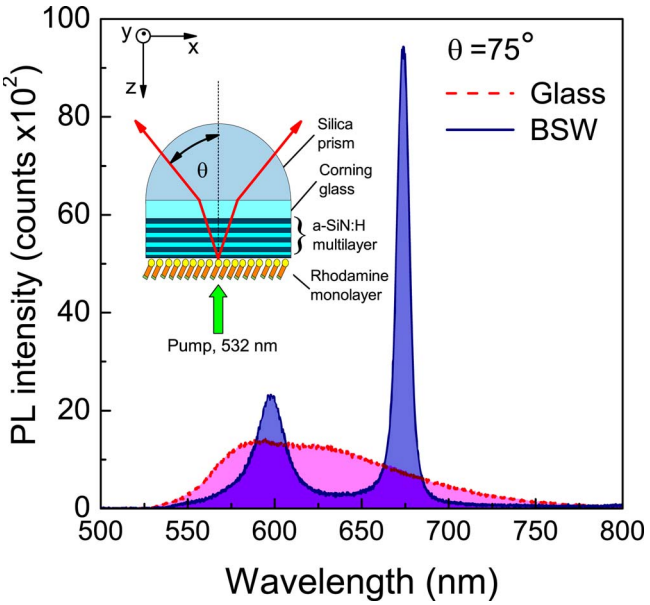


Fig. 2. (Color online) TE-polarized photoluminescence spectra measured at the emission angle $\theta=75°$ from a monolayer of rhodamine bonded to the reference glass substrate (dashed red) and to the a-$Si_{1-x}N_x$:H 1D PhC (solid blue). A sketch of the experimental configuration is also shown.

glass substrate following an established procedure [23].

Angle-resolved photoluminescence (PL) was measured by exciting the rhodamine molecules with a 532 nm cw Nd:YAG laser and collecting the emitted light in the Kretschmann configuration through a silica hemisphere index matched to the sample substrate (see the sketch in Fig. 2). The TE-polarized emitted light was then sent to a grating spectrometer coupled to a LN2-cooled Si CCD. The emission spectra of the rhodamine monolayer bonded to the a-$Si_{1-x}N_x$:H multilayer and to the reference glass substrate, measured at $\theta=75°$, are shown in Fig. 2.

While the monolayer bonded to the reference glass substrate displays the typical rhodamine emission, a remarkable modification of the PL spectrum is observed when the molecules are bonded to the a-$Si_{1-x}N_x$:H 1D PhC. In this last case, two peaks show up at 600 nm and 670 nm. By considering the BSW dispersion relation shown in Fig. 1, we associate the strongest peak at 670 nm with the BSW wave, which is characterized by an intense electromagnetic field at the PhC surface; it therefore promotes a strong interaction with the rhodamine monolayer. The second peak corresponds to the excitation of the Bloch mode associated with the high-energy photonic band edge. Note that here we are not dealing with a guided mode, as there is no light confinement on the substrate side. The different intensity of the two peaks is due to the different quality factors of the two resonances, and the different field distributions. Indeed, in the case of the band-edge mode, the field distribution is peaked in the center of the multilayer. By evaluating the integral of the two PL spectra of Fig. 2 we obtain almost the same value, thus indicating that, in this case, the PhC effect is mainly a redistribution of the emitted light.

In the lower panel of Fig. 3, we show the theoretical and experimental directional enhancements of the emission, defined as the emission intensity of the rhodamine monolayer bonded to the PhC divided by that of the rhodamine monolayer bonded to the reference glass substrate, at a given collection angle $\theta$. When the emitter is coupled to the BSW, we measure a directional enhancement of more than 13. On the other hand, in the PBG at wavelengths between 600 nm and 675 nm there is a substantial suppression of the light emission.

To calculate the rhodamine emission intensity we use a Green function formalism [4] to evaluate the far-field intensity per solid angle, normalized to the Larmor rate. The molecules are modeled as randomly oriented emitting dipoles located at a distance of 1.5 nm from the surface, consistent with the monolayer thickness [23]. We observe a good agreement between the experimental and theoretical curves. Since we neglect scattering and absorption in the theoretical calculation, the theoretical enhancement due to the BSW is larger, and the emission linewidth narrower, than what is experimentally observed. Similarly, the calculated suppression within the stop band is stronger than what is observed.

The corresponding experimental and theoretical attenuated total reflectance (ATR) spectra at the incidence angle $\theta=75°$, in the Kretschmann configura-

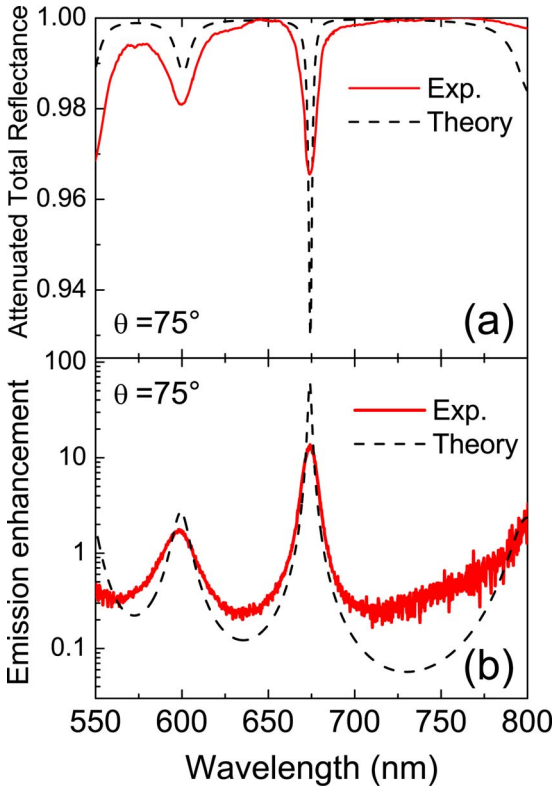


Fig. 3. (Color online) (a) Experimental (solid) and theoretical (dashed) TE-polarized ATR spectra as function of the photon energy for an incidence angle $\theta=75°$. (b) Experimental (solid) and theoretical (dashed) TE-polarized photoluminescence directional enhancement factor as a function of the photon energy at the emission angle $\theta=75°$.

tion, are shown in the upper panel of Fig. 3. Spectral- and angle-resolved ATR experiments allow us to investigate the effects of light localization and the guided-mode properties [24]. As the emission into resonant field structures is enhanced [Fig. 3(b)], so are the losses due to scattering and material absorption when the light is localized in the multilayer in an ATR experiment, leading to dips in the reflectance [Fig. 3(a)]. We calculated the ATR spectrum using a standard transfer matrix algorithm [25], where we introduced a fictitious imaginary part $\varepsilon_2 = 10^{-4}$ in the dielectric function $\varepsilon$ (where $\varepsilon = \varepsilon_1 + i\varepsilon_2$) of the a-$Si_{1-x}N_x$:H to describe the system losses. Although this is a rather crude approximation, we observe a good agreement between the position of theoretical and experimental dips. Note that the small value of $\varepsilon_2$ adopted here does not significantly affect the position of the modes.

In addition, we were able to map the BSW mode dispersion by means of an energy- and angle-resolved PL measurement. The results are shown in Fig. 4, where some of the PhC modes are clearly identifiable. The most intense one corresponds to the BSW, while the other two are associated with the Bloch modes of the PhC structure that exist outside the PBG. From the data in Fig. 4 it is evident that a proper design of the BSW mode dispersion, matched to a specific dye emission, allows us to obtain a field enhancement over a wide spectral and angular range. This may be exploited in an experimental configuration where pump as well as emission wavelengths are both resonant with the BSW.

In conclusion, we observed a directional enhancement of the emission by more than 1 order of magnitude when a rhodamine monolayer was coupled to a BSW in a 1D PhC. The enhancement is associated with the resonant coupling of the emitter to the BSW and results from the strong field confinement near the surface of the structure. Good agreement is found between theory and experiment. These results demonstrate that the use of BSWs, with highly customizable dispersion relations in 1D systems, may become an important feature of efficient light-emitting devices and of optical sensors based on fluorescence.

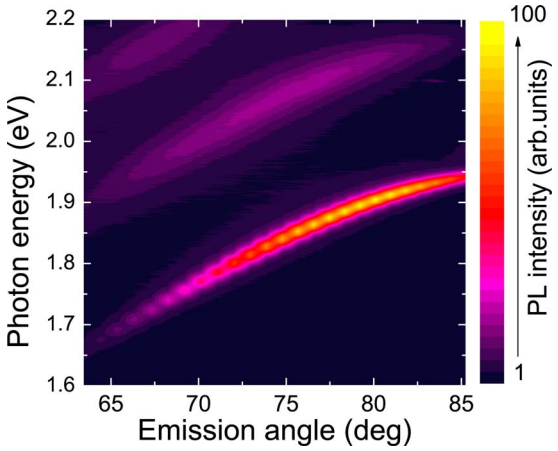


Fig. 4. (Color online) Measured TE-polarized PL of the rhodamine monolayer bonded to the a-$Si_{1-x}N_x$:H as a function of the emission angle and photon energy.

We acknowledge financial support from Fondazione Cariplo and Regione Lombardia (project REGLOM16). This work was also supported by the Ontario Centres of Excellence and by Axela, Inc.

## References

1. P. N. Prasad, *Introduction to Biophotonics* (Wiley, 2003).
2. S. Wedge, J. A. E. Wasey, W. L. Barnes, and I. Sage, Appl. Phys. Lett. **85**, 182 (2004).
3. R. E. Benner, R. Dornhausa, and R. K. Chang, Opt. Commun. **30**, 145 (1979).
4. J. E. Sipe, Surf. Sci. **105**, 489 (1981).
5. J. D. Joannopoulos, S. G. Johnson, J. N. Winn, and R. D. Meade, *Photonic Crystals: Molding the Flow of Light*, 2nd ed. (Princeton U. Press, 2008).
6. E. Yablonovitch, Phys. Rev. Lett. **58**, 2059 (1987).
7. S. Fan, P. R. Villeneuve, J. D. Joannopoulos, and E. F. Schubert, Phys. Rev. Lett. **78**, 3294 (1997).
8. M. Fujta, S. Takahashi, Y. Tanaka, T. Asano, and S. Noda, Science **308**, 1296 (2005).
9. M. Galli, A. Politi, M. Belotti, D. Gerace, M. Liscidini, M. Patrini, L. C. Andreani, M. Miritello, A. Irrera, F. Priolo, and Y. Chen, Appl. Phys. Lett. **88**, 251114 (2006).
10. J. J. Wierer, A. David, and M. M. Megens, Nat. Photonics **3**, 163 (2009).
11. P. Yeh, A. Yariv, and A. Y. Cho, Appl. Phys. Lett. **32**, 104 (1978).
12. A. Yariv and P. Yeh, *Optical Waves in Crystals* (Wiley, 2003).
13. R. D. Pechstedt, P. St. J. Russell, T. A. Birks, and F. D. Lloyd-Lucas, J. Opt. Soc. Am. A **12**, 2655 (1995).
14. R. D. Pechstedt and P. St. J. Russell, J. Lightwave Technol. **14**, 1541 (1996).
15. B. J. Lee, C. J. Fu, and Z. M. Zhang, Appl. Phys. Lett. **87**, 071904 (2005).
16. M. Shin and W. M. Robertson, Sens. Actuators B **105**, 360 (2005).
17. E. Guillermain, V. Lysenko, and T. Benyattou, J. Lumin. **121**, 319 (2006).
18. M. Liscidini and J. E. Sipe, Appl. Phys. Lett. **91**, 253125 (2007).
19. E. Descrovi, F. Frascella, B. Sciacca, F. Geobaldo, L. Dominici, and F. Michelotti, Appl. Phys. Lett. **91**, 241109 (2007).
20. M. Liscidini, M. Galli, M. Patrini, R. Loo, C. Goh, C. Ricciardi, F. Giorgis, and J. E. Sipe, Appl. Phys. Lett. **94**, 043117 (2009).
21. M. Liscidini and J. E. Sipe, J. Opt. Soc. Am. B **26**, 279 (2009).
22. C. Ricciardi, V. Ballarini, M. Galli, M. Liscidini, L. C. Andreani, M. Losurdo, G. Bruno, S. Lettieri, F. Gesuele, P. Maddalena, and F. Giorgis, J. Non-Cryst. Solids **352**, 1294 (2006).
23. P. Pallavicini, G. Dacarro, M. Galli, and M. Patrini, J. Colloid Interface Sci. **332**, 432 (2009).
24. M. Galli, M. Belotti, D. Bajoni, M. Patrini, G. Guizzetti, D. Gerace, M. Agio, L. C. Andreani, and Y. Chen, Phys. Rev. B **70**, 081307 (2004).
25. J. E. Sipe, J. Opt. Soc. Am. B **4**, 481 (1987).