Abstract. We model the modification in the spontaneous emission (SE) rate of an emitting dipole placed on the surface of a two-dimensional (2D) L3 optical nanocavity using 3D FDTD calculations. We show that by introducing a concave region into the cavity-surface, the cavity $Q$-factor is largely unaffected, however the electromagnetic field intensity at the dielectric–air interface is significantly enhanced compared to a regular L3 (planar) structure. Our calculations indicate that such a modification of the cavity surface leads to peak emission intensity enhancements (at the cavity-mode resonance) for a dipole emitter of 0.1 nm linewidth by a factor of 33 times. For an emitter having a linewidth of 12 nm, a peak intensity enhancement of 43 times is predicted. Finally, we show that enhancements of up to 42 times in SE rate can be expected for surface-bound (organic) emitters, with our structures having possible applications as nano-sensor devices and single-photon light-sources.
Photonic crystal (PC) nanocavity structures [1]–[7] are able to confine light within a mode volume \( V \) of the order of \((\lambda/\eta)^3\). An important measure of the effectiveness by which light is confined in the cavity is the cavity Q-factor defined as \( Q = E/\Delta E \), where \( E \) is the energy of the confined photons and \( \Delta E \) is the energy linewidth of the homogeneously broadened optical mode. Nanocavities have been demonstrated having Q-factors of up to \( 10^6 \) for membrane structures based on Si [8] and 20 000 for GaAs-based structures containing quantum dots [9, 10]. Such cavities are an attractive system in which to study fundamental physical processes such as the Purcell effect [11, 12], which describes the enhancement or suppression of the spontaneous emission (SE) rate of an emitting dipole placed in an optical cavity \((\Gamma')\) compared to its free-space value \((\Gamma_0)\). Here, the modified optical density of states enhances or suppresses the SE rate as given [6, 12] by

\[
\frac{\Gamma}{\Gamma_0} = F_{\text{cav}} \left( \frac{E(r) \cdot \mu}{|E_{\text{max}}| \cdot |\mu|} \right)^2 \left( 1 + 4Q^2 \left( \frac{\lambda_e}{\lambda_{\text{cav}}} - 1 \right)^2 \right)^{-1} + F_{\text{PC}},
\]

where \( \lambda_e \) is the emission wavelength of the dipole, \( \lambda_{\text{cav}} \) is the cavity resonance wavelength, \( E(r) \) is the local electric field experienced by the emitting dipole, \( \mu \) is the electric dipole moment of the emitter, \( E_{\text{max}} \) is the maximum amplitude of the electric field and \( F_{\text{cav}} \) is the cavity Purcell factor expressed by \( F_{\text{cav}} = 3Q\lambda_{\text{cav}}^3/(4\pi^2n^3V) \) (where \( n \) is the refractive index of the nanocavity dielectric medium). A term \( F_{\text{PC}} = \Gamma_{\text{PC}}/\Gamma_0 \) is also included to account for suppression of SE resulting from the photonic band-gap of the PC in the absence of a cavity [6]. From equation (1), it is clear that to maximize the Purcell effect, the cavity has to have a high \( Q/V \) ratio, with the dipole emitter having a high spatial and spectral overlap with the electromagnetic field in the cavity.

The Purcell effect has been investigated intensively in materials such as GaAs [6, 7, 10], [12]–[18]. This material system has two main advantages: firstly, the high refractive index of GaAs can be used to create nanocavities having a high Q-factor [9, 10]. Secondly, epitaxially-grown quantum dots can be engineered into a region corresponding to the peak amplitude of the confined electromagnetic field (in the centre of the slab-waveguide) permitting structures with a high Purcell factor to be realized. Such systems have a range of potential applications in photonics and quantum communications, including single photon light sources [7] and ultra low-threshold lasers [5].

There is, however, increasing interest in the design and construction of so-called ‘hybrid-semiconductor’ heterostructures (based on both organic and inorganic materials) [19]–[21] and micro- and nano-photonic structures in which the active emissive material is an organic-chromophore [22]–[25]. Organic-emitters can be processed into unconventional architectures using inexpensive techniques and have the advantage of excitonic emission at room temperature. Indeed, we have recently shown that nanocavities operating at visible-wavelengths can be fabricated based on materials such as TiO\(_2\) (making them suitable for many organic dyes) and are anticipated to have a Q-factor of a few thousand [26]. Our work is partly motivated by the idea of using a nanocavity as a chemical or biochemical sensor device to follow the binding of organic materials onto the cavity surface [27]. However, the optical field amplitude \( E(r) \) of the fundamental mode at the surface of a regular \( L3 \) cavity is less than 30% of the field maximum at the cavity centre. As the Purcell factor is proportional to \( |E(r)|^2 \), this can significantly limit
Figure 1. A schematic of the nanocavity structures investigated is shown in parts (a) and (b). The PC surrounding the nanocavity has a lattice constant of $a = 235 \text{ nm}$, a slab thickness of $d = 0.9a$, a hole radius of $r = 0.3a$ and a refractive index of $n = 2.45$. Part (c) plots the amplitude of the electric field ($E_y$) of the fundamental mode of structure C, calculated in an $(x, z)$-plane through the cavity centre and surrounding PC (see text for details). In the calculations we used the parameters $b = 500$, $c = 250$ and $h = 50 \text{ nm}$.

the maximum enhancement of the SE rate that can be engineered in such surface-emitter based structures.

In this paper, we use a three-dimensional (3D) finite difference time domain (FDTD) approach\(^2\) to theoretically model a straightforward modification to an $L3$ cavity \[^3\] that we predict will significantly enhance the Purcell effect of an emitting dipole placed on a nanocavity surface. Importantly, all our simulations are based on the use of TiO$_2$ as the cavity dielectric medium. This material combines relatively high refractive index ($n = 2.45$) and very low absorption over the whole of the visible electromagnetic spectrum, making it an ideal system to be combined with a range of fluorescent organic materials. Our structures are thus a good example of how both organic and inorganic materials could be engineered into a ‘hybrid’ device that displays functionality that is not easily obtainable using either material alone. Figures 1(a) and (b) show a schematic of our design. The structure is based on an $L3$ nanocavity having a concave ellipsoid region defined into the cavity centre. The geometry of the concave surface is defined using the parameters $b$, $c$ and $h$, where $b$ and $c$ are the major and minor axes of an ellipse respectively, and $h$ is the depth of the concave surface (at its centre) defined with respect to the top surface of the nanocavity. The key idea behind this design is to place the dipole emitter in a region of higher electromagnetic field without significantly reducing the cavity $Q$-factor or

\[^2\] The 3D FDTD code (CrystalWave) used in this work is a product of Photon Design Ltd, http://www.photond.com.
Table 1. Structural and optical parameters of the cavities investigated in this work. Note that the origin of the coordinate system \((x, y, z)\) is defined at the centre of the cavity surface.

| Structure | A | B | C | D |
|-----------|---|---|---|---|
| \(b, c, h\) (nm) | 0, 0, 0 | 0, 0, 0 | 500, 250, 50 | 120, 120, 106 |
| Cavity region | Planar | Planar | Concave | Hole |
| Dipole position \((x, y, z)\) | Centre | Surface | Surface | Centre |
| \(\lambda_{	ext{cav}}\) (nm) | 706.3 | 706.6 | 705.4 | 702.9 |
| \(Q\)-factor | 980 | 940 | 880 | 350 |
| 12 nm dipole, EE at resonance | 92 | 7 | 43 | 84 |
| 0.1 nm dipole, EE at resonance | 87 | 4.6 | 33 | Not calculated |

significantly changing the resonance wavelength. Note that the particular design proposed only applies to optical modes having an antinode at the cavity centre. These include the fundamental mode of \(L^3\) cavities or the dipole modes of Lee cavities [28]. Our modification would not apply to structures having a node at the cavity centre such as the hexapole mode of Lee cavities.

2. Results and discussions

We have explored four types of \(L^3\) cavities as summarized in table 1. Structures A and B represent a cavity having a flat surface, with an emissive dipole located in the cavity centre (A) or at the cavity surface (B). Structure C corresponds to a cavity having a relatively wide \((c = 500\text{ nm})\) but shallow \((h = 50\text{ nm})\) ellipsoid defined into its surface. Structure D has a small circular hole etched into the centre of the cavity to access regions of highest confined optical field \((b = c = 120\text{ nm}, h = 106\text{ nm})\). Each structure is defined into the centre of a cavity composed of three linearly aligned missing air holes \((L^3\text{ type})\) with the first nearest-neighbouring holes displaced by a distance of \(\Delta s = 0.21a\) to maximize the \(Q\)-factor. Each cavity is imbedded in a TiO\(_2\) 2D triangular lattice PC slab having a lattice constant of \(a = 235\text{ nm}\), a slab thickness of \(d = 0.9a\), a hole radius of \(r = 0.3a\) and a refractive index of \(n = 2.45\). In all cases, the PC is constructed on a low refractive index substrate having \(n_{\text{sub}} = 1.33\), designed to replicate the refractive-index value of certain optical resists\(^3\) or aerogels.

In our previous work [29], we showed that such PCs are able to support a full TE band gap of width of 80 nm centred at 682 nm with the fundamental optical mode having a cavity \(Q\)-factor of 1000. The fundamental mode of an \(L^3\) nanocavity has an \(E_y\) field distribution localized within the cavity region and consists of three lobes of even parity along both \(x\)- and \(y\)-directions, with a field maximum at the cavity centre, and \(y\)-polarized vertical emission [3, 30]. Figure 1(c) shows the electric field distribution \((E_y)\) of the cavity fundamental mode.

\(^{3}\) For example LS-2233 optical resist manufactured by Lightspan™, LLC and distributed by Polymer Systems Technology Ltd.
in cross-section through the cavity centre. It is clear that the electric field is strongly localized within the cavity having a maximum value at the cavity centre. This indicates that the low index substrate does not significantly reduce the cavity electromagnetic field confinement.

The optical properties of the fundamental mode of the cavities defined in table 1 were modelled using a 3D FDTD approach (see footnote 2). In the calculations the PC size was set at \((50a \times 16\sqrt{3}a)\). Each unit cell was discretized in all three dimensions with a grid spacing of \(\Delta G = a/10\), and a time step of \(0.99\sqrt{3/7}/c\), where \(c\) is the speed of light. Boundary conditions were implemented by introducing perfect matching layers at the boundaries of the structure. In the calculations the emitter was modelled in the form of a classical dipole oriented in the \(y\)-direction and positioned in the centre of the cavity \((x = y = 0)\) at various depths \(z\), with \(z\) defined with respect to the surface of the surrounding PC—see figure 1(b). This emitting dipole is then used to excite the cavity mode. In all cases, we place the emission peak of the dipole in exact resonance with the cavity mode \((\lambda_e = \lambda_{cav})\). Two limiting cases for emissive dipoles were explored. The first had an emissive linewidth of 12 nm (30 meV at 706 nm), replicating the emission spectrum of a single molecule or a single colloidal quantum dot at room temperature. The second case considered a dipole having a linewidth of 0.1 nm (250 \(\mu\)eV at 706 nm), replicating the low temperature emission spectrum of a colloidal quantum dot. Using our FDTD calculations, we are able to extract the \(Q\)-factor of the cavity, the emission integrated within an external cone angle of \(\sim 26^\circ\) which represents the practical collection geometry of a microscope having an objective lens with a numerical aperture of 0.44, and the \((x, y, z)\) electric field components within the cavity structure. In each case, we use the results of our calculations to extract the peak enhancement of emission compared to a dipole emitter in free-space.

We anticipate that the structures that we have modelled will be relatively easy to fabricate as they do not require selective etching processes to create an air bridge structure. For example, the low refractive index layer (e.g. an optical resist) can be deposited from solution onto a suitable substrate using spin coating, whilst the \(\text{TiO}_2\) layer could be deposited on the low refractive index layer using electron-beam ion-assisted deposition. It is possible to pattern such materials to create a PC using one of a variety of techniques including electron beam lithography followed by dry etching or focused ion beam (FIB) lithography. The concave depression formed in the centre of the cavity could similarly be defined using techniques such as FIB lithography. Finally there are a number of methods that could be used to deposit an active emitting material on to the cavity surface including near field optical lithography [31] or dip-pen lithography [32]. Such scanning probe lithography techniques have the advantages of very high spatial resolution and registration, potentially permitting an emitting material to be accurately aligned on the nanocavity surface.

Figure 2(a) shows the calculated emission spectra from structures A, B, C and D containing a 12 nm linewidth dipole. In all cases, each spectrum represents emission ‘collected’ over an external cone angle of \(26^\circ\). Also plotted is the emission spectrum of a reference free-space dipole emitting over the same external cone angle. We summarize the cavity \(Q\)-factor and the emission enhancement (EE) from all the structures in table 1. We define the EE as the peak emission intensity at resonance determined from the nanocavity (integrated over a cone angle of \(26^\circ\)) divided by the peak emission intensity of a free-space dipole (integrated over the same angle). It can be seen that the regular cavity structure A in which the dipole is embedded in the centre of the cavity layer has the highest \(Q\)-factor of 980 and also the largest EE at the peak of the cavity resonance of 92 times. Although introducing a concave region to the cavity centre (structure C) results in a blue shift by of 0.9 nm in the position of the cavity fundamental mode,
Figure 2. Calculated emission spectra for structures A, B, C and D as detailed in table 1. Part (a) shows the result of calculations assuming a 12 nm linewidth emitting dipole. Part (b) shows calculated spectra assuming a 0.1 nm linewidth emitting dipole. Note that, in each case, we have arranged the emissive dipole to be in direct resonance with the fundamental cavity mode (i.e. $\lambda_e = \lambda_{cav}$).

In structure D, we explore the effect of etching a 120 nm diameter hole into the centre of the cavity slab waveguide to couple the dipole to the maximum of the electromagnetic field associated with the cavity fundamental mode. It can be seen however that this structure reduces the $Q$-factor of the cavity by almost 3 times. Nevertheless, the effect of the high electromagnetic field experienced by the 12 nm linewidth dipole results in a peak emission intensity that is within 90% of that achieved by placing the emission dipole in the centre of a regular planar cavity (structure A). As we discuss below, such large enhancements in EE from low $Q$-factor cavities are particularly interesting for applications where the emissive dipoles are anticipated to undergo time-dependent spectral shifts.

Figure 2(b) shows the calculated integrated emission spectra from structures A, B, C and D containing a 0.1 nm linewidth dipole. Here we find that when we place the dipole at the centre of a planar cavity, the peak emission intensity is enhanced by 87 times compared to its free-space value—a value similar to that obtained with a 12 nm dipole. It is again apparent that emission from the narrow line-width dipole is significantly enhanced by placing it on the surface of the cavity having a concave surface (structure C) compared to the simple planar structure (structure B). This again confirms that structuring the surface of a regular $L3$ cavity to increase the electromagnetic field amplitude at the dielectric–air interface is a very promising method to enhance the peak emission intensity from a suitable surface-bound dipole. As we demonstrate below, this approach is anticipated to result in a significant increase in SE rate of a surface-bound organic chromophore or colloidal quantum-dot.
These results therefore suggest that we should expect an enhancement in peak emission intensity from a dipole located in all the structures modelled compared to that from a dipole in free-space. Clearly there are two mechanisms that can result in such an enhancement: (i) an enhancement in SE rate, and (ii) an enhancement in optical extraction efficiency (i.e. a redirection of light towards the detector). At present, our FDTD simulations are not able to easily distinguish between these two possibilities, however simplistic symmetry arguments would suggest that both structures B and structure C will have similar extraction efficiencies, as both structures have an emitting dipole placed at an air–dielectric interface. The results presented in table 1 show however that the EE from structure C is 6 times higher than that of structure B, suggesting that a significant contribution of the enhancement results from the Purcell effect.

We can however also use equation (1), together with the FDTD calculated values of the cavity Q-factor and the electromagnetic field distribution to estimate the enhancement in the SE rate from the various structures detailed in table 1. In this calculation, we implicitly assume that \( \mu \) is aligned with the \( y \)-direction (the polarization of the cavity mode). Of course, in a practical embodiment of this structure, \( \mu \) would be free to take any orientation, and thus the SE enhancements that we determine here should be considered as maximum possible values. Note that our FDTD calculations suggest that the term included to account for a suppression of SE resulting from the photonic band-gap of the PC in the absence of a cavity (\( F_{PC} \)) is equal to 0.02 within the band gap region.

Figure 3 shows the spatial distribution of the SE rate enhancement for a dipole in structures A, B and C at resonance (i.e. \( \lambda_e = \lambda_{cav} \)). From the results in figure 3, it can be seen that SE rate enhancement is a strong function of space and it takes its maximum value close to the \((x, y)\) centre of the cavity as expected. When the dipole emitter is embedded within the centre of a planar cavity (structure A), we predict a maximum enhancement in SE rate of 82 times compared to a free-space dipole. This is reduced to 10 times the free-space emission-rate when the dipole is placed on the surface of a planar cavity (structure B) resulting from the lower electromagnetic field found at the air–dielectric interface. However the structure having the shallow, concave-surface (structure C) has an enhancement in SE rate anticipated to be almost 42 times that of its free-space rate, again illustrating the potential benefit of our proposed cavity modification.

In the above analysis, we did not take into account any possible spectral mismatch between the emitting dipole and the cavity resonance, which is expressed by the term

\[
\left( 1 + 4Q^2 \left( \frac{\lambda_e}{\lambda_{cav}} - 1 \right)^2 \right)^{-1}
\]

in equation (1) (i.e. we set \( \lambda_e = \lambda_{cav} \)). This approach is applicable for the case of a broad emitting source, where we are mainly concerned with the SE rate enhancement at resonance. For the case of a narrow linewidth source (replicating a dipole at low temperature), it is important to have a large spectral overlap between the emitter and the cavity mode. For the cavities investigated in this work where we assume a dipole having a linewidth (full width at half maximum) of 0.1 nm (equivalent to 250 \( \mu eV \) at 706 nm) the spectral mismatch factor drops from unity at resonance, to 0.50 when \( \lambda_e = \lambda_{cav} \pm \Delta \lambda_{cav}/2 \) (with \( \Delta \lambda_{cav} \) being the cavity mode linewidth). For structure C, we calculate that \( \Delta \lambda_{cav} \approx 0.8 \) nm (equivalent to \( \sim 2 \) meV for \( \lambda_{cav} = 705 \) nm). This therefore defines the range over which a (time-dependent) spectral shift of dipole emission energy could

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occur without resulting in more than a factor of two reduction in SE rate. In general, this linewidth is comparable with the energy-shifts resulting from spectral-diffusion measured at low temperature in materials such as CdSe colloidal quantum dots (between 2 and 4 meV) [33] and single terylenediimide molecules (between 2 and 10 meV) [34]. This relatively high degree of tolerance against spectral-diffusion suggests that our proposed structures combining relatively low $Q$-factors with high SE rate enhancement also offer promise as single-photon light-sources for quantum-communication systems.

Figure 3. The SE rate enhancement calculated as a function of position in the $x$–$y$-plane for structure A at $z = 106$ nm (cavity centre), structure B at $z = 0$ (cavity surface) and structure C at $z = 50$ nm (corresponding to the lowest point on the concave cavity surface).
3. Conclusions

In summary, we have modelled a straightforward modification of the structure of an $L3$ nanocavity to improve the SE rate of an emitting dipole placed onto the cavity surface. Our modification involves creating a cavity having a concave surface, permitting us to place the emitting dipole closer to the electric field antinode of the fundamental mode of the cavity. The enhanced electromagnetic field experienced by the dipole is anticipated to increase the Purcell factor (the enhancement in SE rate), without significantly reducing the cavity $Q$-factor. Our calculations showed that in the case of a dipole source of 12 nm linewidth (representative of the emission from a single molecule or a single quantum dot at room temperature), the peak emission intensity is enhanced by a factor of 43 times. Similarly large enhancements can also be achieved when the linewidth of the emissive dipole is significantly narrower than that of the cavity mode. For example, in the case of a narrow emission source of 0.1 nm linewidth (representative of a single quantum dot at cryogenic temperature) in a cavity of mode linewidth of 0.7 nm, we predicted a peak intensity enhancement by a factor of 33 times. By using our calculated values of cavity $Q$-factor and electromagnetic field amplitude, we showed that a suitably aligned dipole placed on the surface of a modified cavity might have its SE rate enhanced by a factor of $\sim 42$ times compared to a free-space dipole emitter.

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