Optimal trapping of monochromatic light in designed photonic multilayer structures

Fabian Spallek, Andreas Buchleitner and Thomas Wellens

Albert-Ludwigs-Universität Freiburg, Physikalisches Institut, Hermann-Herder-Str. 3, D-79104 Freiburg i. Br. Germany

E-mail: fabian.spallek@physik.uni-freiburg.de

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Abstract
We devise an optimised bi-component multi-layered dielectric stack design to enhance the local irradiance for efficient photovoltaic upconversion materials. The field intensity profile throughout the photonic structure is numerically optimised by appropriate tuning of the individual layers’ thicknesses. The optimality of the thus inferred structure is demonstrated by comparison with an analytically derived upper bound. The optimised local irradiance is found to increase exponentially with the number of layers, its rate determined by the permittivity ratio of the two material components. Manufacturing errors which induce deviations from the optimised design are accounted for statistically, and set a finite limit to the achievable enhancement. Notwithstanding, realistic assumptions on manufacturing errors still suggest achievable irradiances which are significantly larger than those obtained with the recently proposed Bragg stack structures.

Keywords: upconversion, photonic multilayer structures, photon management, light trapping, optimal design

1. Introduction

Photon upconversion, i.e., the conversion of two photons with smaller energy into one photon with larger energy, offers promising possibilities to improve the efficiency of solar cells [1–3]. The majority of currently used photovoltaic technologies is based on materials with semiconductor properties, where a certain minimum energy per photon (defined by the bandgap of the semiconductor) is required to create an electron–hole pair. Photons with smaller energies are not absorbed by the solar cell, and are therefore lost for the purpose of light-energy conversion. In the case of silicon, the resulting loss amounts to approximately 20% of the power of the incident solar radiation [4].

In principle, these losses can be reduced by shifting the energy of the transmitted photons above the bandgap, thereby rendering them usable for the solar cell. Experimentally, a thus achieved relative increase of the current generated by a silicon solar cell of about 0.55% has already been demonstrated [5]. This increase was realised by placing an upconverter material made of monocristalline BaYF$_3$: 30% Er$^{3+}$ on the non-irradiated side of the solar cell. In order to further increase the upconversion efficiency, it was proposed to embed the upconversion material within suitably chosen photonic structures [6, 7]. In principle, such structures may influence the upconversion luminescence in different ways: first, they can be used in order to enhance the local irradiance of the upconversion material as compared to the non-concentrated incident light. This enhancement also increases the upconversion luminescence due to the nonlinearity of the underlying process [8]. Second, photonic structures change the local density of states and, consequently [9], the rates of spontaneous emission processes occurring within the upconverter ions. Recently, a Bragg stack [10] consisting of layers with periodically alternating refractive indices has been identified as a particularly promising candidate of a photonic structure, which, according to the results of numerical simulations, leads to a considerable enhancement of the upconversion luminescence for experimentally realistic parameters [7].

According to the design proposed in [7], the thicknesses of all layers forming the Bragg stack are determined by a single parameter (the so-called design wavelength $d$, see section 3.2 below), which is then optimised in order to maximise the upconversion luminescence. In the present paper, we study to which extent such structures can be further improved by optimising the thickness of each constituent layer individually, such
as to maximise the local irradiance at a given position. We design a photonic structure consisting of dielectric layers with given permittivities such that, for an incident monochromatic plane wave, the intensity of the electric field inside the photonic structure is maximised. In other words, we aim at trapping the incident photons inside the photonic structure for a time as long as possible, by the creation of strongly localised, narrow resonance eigenmodes which result from the boundary conditions as set by the permittivity landscape. In contrast to traditional methods of light trapping [11, 12] based on principles of geometric optics, our optimisation strategy relies on appropriately tuned interferences between multiply reflected wave amplitudes, and is therefore restricted to waves with a given wavelength and angle of incidence. From a more general perspective, our problem to define the optimal photonic structure is closely related to problems of photon management [13] and of network design for optimised excitation transport in light harvesting units made by nature [14–17].

As already mentioned above, our optimised design—in contrast to the Bragg structures proposed in [7]—exhibits different thicknesses for each individual layer. Finding the optimal structures for a large number of layers (up to $N \approx 30$) therefore requires extensive numerical optimisation. The optimality of our numerically obtained structures is confirmed by comparison with an analytical upper bound. Moreover, we also take into account fabrication errors and determine structures which are especially robust with respect to these errors.

The paper is organised as follows: in section 2, we introduce the Helmholtz equation for the electric field at a given position inside a one-dimensional multi-layered photonic structure, describe the transfer matrix method as an efficient tool for its solution and outline the numerical algorithm to determine optimised structures in which the intensity of the electric field is maximised. In section 3, we derive analytical upper bounds for the achievable intensity enhancement, compare them to the characteristic properties of our numerically optimised structures. We analyse the effect of fabrication errors as well as a finite spectral bandwidth of the incident light leading to a second class of robust optimised structures. In section 4, we summarise and discuss our results, and give perspectives for future studies.

2. Model and methods

We consider a stationary scattering scenario for a light field of given polarisation, fixed frequency $\omega$, and wave vector normal to the photonic structure’s surface. The time dependence of the electric field is separated as

$$E(\vec{r}, t) = \mathcal{E}(\vec{r}) e^{-i\omega t},$$

and Maxwell’s equations (without sources) can be condensed into the Helmholtz equation for the space-dependent electric field component $\mathcal{E}(\vec{r})$:

$$\nabla^2 \mathcal{E} + \frac{\omega^2}{c^2} \varepsilon(\vec{r}) \mathcal{E} = 0.$$

Given a one-dimensional multilayer structure with layers in the $x$–$y$ plane, and homogeneous dielectric materials defined by a piecewise constant permittivity $\varepsilon(\vec{r}) = \varepsilon(z) = \varepsilon_n$ in the $n$th layer, and without absorption, i.e., $\varepsilon(z) \in \mathbb{R}$, we are left with the one-dimensional Helmholtz equation:

$$\frac{\partial^2 \mathcal{E}_n}{\partial z^2} + \frac{\omega^2}{c^2} \varepsilon_n \mathcal{E}_n = 0 \quad (3)$$

in layer $n \in \{1, \ldots, N\}$. The general solution of (3) reads $\mathcal{E}_n(z) = A_n e^{ik_n z} + B_n e^{-ik_n z}$, with wave number $k_n = \frac{\omega}{c} \sqrt{\varepsilon_n}$ and amplitudes $A_n$ and $B_n$ for right- (and left-) propagating waves.

The multilayer structure consists of two alternating materials, one with permittivity $\varepsilon_{\text{high}}$ containing the upconverter, and a second one with larger permittivity $\varepsilon_{\text{low}}$ providing the photonic structure. We consider an odd number $N$ of layers, with permittivities $\varepsilon_n = \varepsilon_{\text{high}}$ for odd, and $\varepsilon_n = \varepsilon_{\text{low}}$ for even $n$, respectively. The $n$th layer is confined between $z_{n-1}$ and $z_n$, with $z_n > z_{n-1}$ and $z_0 = 0$. The stack is embedded in air, such that $z_0 = 1$ for $z < 0$ and $z_{N+1} = 1$ for $z > z_N$. The stack is thus defined by the layer thicknesses $d_n = z_n - z_{n-1}$ and the associated values of $\varepsilon_{\text{high}}$ and $\varepsilon_{\text{low}}$, see figure 1.

2.1. Transfer matrix method

The general solution of (2) for a one-dimensional structure as depicted in figure 1 reads:

$$\mathcal{E}_n(z) = \sum_{n=1}^{N} (A_n e^{ik_n z} + B_n e^{-ik_n z}) \Theta(z - z_{n-1}) \Theta(z_n - z)$$

$$+ (A_0 e^{ik_0 z} + B_0 e^{-ik_0 z}) \Theta(-z)$$

$$+ (A_{N+1} e^{ik_{N+1} z} + B_{N+1} e^{-ik_{N+1} z}) \Theta(z - z_N),$$

with amplitudes $A_n$ and $B_n$ related through the boundary conditions at the layers’ interfaces. Maxwell equations in non-magnetic media without external sources enforce the continuity of $\mathcal{E}_n$ as well as of $\partial \mathcal{E}_n$ at the position $z_n$ of every interface, i.e.:

$$A_n e^{ik_n z_n} + B_n e^{-ik_n z_n} = A_{n+1} e^{ik_{n+1} z_n} + B_{n+1} e^{-ik_{n+1} z_n},$$

$$k_n A_n e^{ik_n z_n} - k_n B_n e^{-ik_n z_n} = k_{n+1} A_{n+1} e^{ik_{n+1} z_n} - k_{n+1} B_{n+1} e^{-ik_{n+1} z_n}.$$  

This relates the amplitudes $v_n = (A_n, B_n)^T$ and $v_{n+1} = (A_{n+1}, B_{n+1})^T$ within two adjacent layers $n$ and $n + 1$ according to

$$v_n = M_n \cdot v_{n+1},$$

with the transfer matrix [18]

$$M_n = \frac{1}{2k_n} \left[ \begin{array}{cc} (k_n + k_{n+1}) e^{ik_{n+1} z_n} & (k_n - k_{n+1}) e^{-ik_{n+1} z_n} \\ (k_n - k_{n+1}) e^{ik_{n+1} z_n} & (k_n + k_{n+1}) e^{-ik_{n+1} z_n} \end{array} \right].$$

Note that the $M_n$ is fully determined by the geometry encoded in $\varepsilon(z)$, as it only depends on the positions $z_n$ of the surfaces between the layers, and on the wave vectors $k_n = \frac{\omega}{c} \sqrt{\varepsilon_n}$. The amplitude $A_0$ of the incoming wave is normalised such
Figure 1. Photonic structure consisting of alternating layers with permittivities $\varepsilon_{\text{high}}$ and $\varepsilon_{\text{low}}$, respectively. The amplitudes of the right- and left-propagating waves inside layer $n$ (with thickness $d_n = z_n - z_{n-1}$) are denoted by $A_n$ and $B_n$, respectively. The incident wave with normalised amplitude $A_0 = 1$ arrives from the left-hand side.

that $A_0 = 1$. Because of possible reflection at $z = 0$, it follows that $B_0 = 0$ and, since there is no incoming wave travelling in negative $z$-direction from $z > z_N$, we have $B_{N+1} = 0$.

The reflection coefficient of the full structure is given by $R = |B_0|^2$ while the transmission reads $T = |A_N+1|^2$. From energy flux conservation, it follows that $R + T = 1$ and

$$\sqrt{\varepsilon_n} (|A_n|^2 - |B_n|^2) = T$$

for each layer $n$.

The solution of (4), given by the amplitudes $v_n = (A_n, B_n)^T$, $n = 1, \ldots, N$, can now be determined by propagation of the initial condition $(A_{N+1}, B_{N+1}) = (1, 0)$ across the multilayer structure, by iterative application of the transfer matrix $M_n$: $(A_n, B_n)^T = M_n \cdot (A_{n+1}, B_{n+1})^T$. Once the input face of the structure is reached, $(A_0, B_0)^T = (0, 1)^T$, in a final step all amplitudes $A_n$, $B_n$ are renormalized, i.e. $A_n = \frac{A_n}{\varepsilon_n}$ and $B_n = \frac{B_n}{\varepsilon_n}$, such that $A_0 = 1$.

2.2. Numerical optimisation

To improve the efficiency of the upconversion process, we seek to maximise the field intensity within the upconversion layers of the photonic structure. The corresponding target function is given by

$$\gamma = \frac{1}{\varepsilon_n} \sum_{m=1}^{z_2} \left( \sum_{m=1}^{z_m-1} \int_{z_{m-1}}^{z_m} dz \right) I(z),$$

which quantifies the average intensity in the upconversion volume of the multilayer device, and owes its name to the fact that $I$ is normalised to the intensity of the incoming wave. At least at weak irradiation, the upconversion efficiency is expected to scale quadratically with the intensity [8]. One may therefore alternatively average over $I^2(z)$ in (10) rather than over $I(z)$. However, we verified that our subsequent results are essentially insensitive to such a replacement.

Note that, due to the periodic intensity modulation within each layer $n$ given by

$$I(z) = |A_n|^2 + |B_n|^2 + 2 \Re(A_n B_n^* e^{i2k_n z})$$

which is a direct consequence of the superposition of right- and left-running amplitudes $A_n$ and $B_n$, respectively, it suffices to optimise $d_n$ in the range $d_{\text{min}} < d_n < \frac{\lambda}{2} - d_{\text{min}}$ with $\lambda = 2\pi/k_n$ and $d_{\text{min}} = 0.025\lambda_n$ to exclude vanishing layer thicknesses as output of our optimisation procedure.

The optimal thickness profile $d_n$, $n = 1, \ldots, N$, is then numerically inferred through the following iterative procedure:

(i) Insert two additional layers with thickness $d_{N+1} = d_{N+2} = \frac{\lambda}{2}$ in the middle of the stack of a given optimised structure with $M = 2$ layers, with these previously given $M - 2$ layers’ thicknesses unchanged.

(ii) Optimise this $M$-layer structure with the downhill simplex method of Nelder and Mead [19, 20].

(iii) From the analytical benchmark (24), we expect that $\gamma$ increases exponentially as a function of $M$, i.e. $\gamma(M) = |\gamma(M - 2)|^2 / \gamma(M - 4)$. Verify whether the result of (ii) leads to an enhancement $\gamma$ which agrees (up to a relative error not larger than 2%) with this expectation.

- If agreement is given, continue with (i) until $M = N$.
- If not, re-initialise the downhill simplex algorithm with statistically perturbed initialised values of $d_{N+1}$ and $d_{N+2}$.

3. Results

Before we discuss specific properties of the numerically optimised structures, let us first derive an analytical upper bound for the achievable maximal local intensity $I(z)$ at a given position $z$ within the photonic structure, given the fundamental set of equations (4)–(11). As we will see, the upper bound for $R(z)$ increases exponentially as a function of the number of layers which separate the position $z$ from the left or the right boundary of the photonic structure, respectively. From this, we estimate an upper bound for the integrated local intensity, which defines the intensity enhancement factor $\gamma$ as given in equation (10). Also $\gamma$ increases exponentially as a function of the total number $N$ of layers. The above bound enters as a benchmark into the numerical optimisation procedure as described in 2.2 further up, and in addition provides useful physical insight.

3.1. Analytical upper bound

To derive an upper bound for the intensity that can, in principle, be reached with a given number $N$ of layers, we first observe that, according to (11), the intensity $I(z)$ inside layer $n$ fulfills:

$$I(z) \leq (|A_n|^2 + |B_n|^2) = I_0 + \sqrt{I_0^2 - \frac{T^2}{\varepsilon_n}},$$

where we used flux conservation, see (9), and introduced the background intensity

$$I_0 = |A_n|^2 + |B_n|^2$$

which can be expected to scale quadratically with the intensity $I(z)$. At least at weak irradiation, the upconversion efficiency is expected to scale quadratically with the intensity $I(z)$.
inside layer \( n \), defined as the constant (i.e. non-oscillating) term in the expression (11) for \( I(\zeta) \). Since \( v_n = (A_n, B_n)^T \), the background intensity \( I_n \) can be interpreted as the squared norm of the vector \( v_n \in \mathbb{C}^2 \) induced by the standard scalar product in \( \mathbb{C}^2 \), i.e., \( I_n = v_n^\dagger \cdot v_n \). According to (7), we have:

\[
v_n^\dagger \cdot v_n = \sum_{n=0}^{N+1} a_{n+1}^* b_n \cdot \bar{a}_n b_{n+1}.
\]

Therefore, the ratio \( I_n / I_{n+1} = (v_n^\dagger, v_{n+1}) / (v_{n+1}^\dagger, v_{n+1}) \) of the background intensities in adjacent layers is bounded between the two eigenvalues of the Hermitian matrix:

\[
M_n^\dagger \cdot M_n = \frac{1}{2 k_n} \begin{pmatrix} k_n^2 + k_{n+1}^2 & \left(k_n^2 - k_{n+1}^2\right) e^{-2i k_n \cdot \zeta_n} \\ \left(k_n^2 - k_{n+1}^2\right) e^{2i k_n \cdot \zeta_n} & k_n^2 + k_{n+1}^2 \end{pmatrix}.
\]

The eigenvalues and eigenvectors of this matrix are:

\[
\lambda_n^{(1)} = 1, \quad w_n^{(1)} = \frac{1}{\sqrt{2}} \begin{pmatrix} 1, \ e^{2i k_n \cdot \zeta_n} \end{pmatrix},
\]

\[
\lambda_n^{(2)} = \left(k_n - k_{n+1}\right)^2, \quad w_n^{(2)} = \frac{1}{\sqrt{2}} \begin{pmatrix} 1, -e^{2i k_n \cdot \zeta_n} \end{pmatrix}.
\]

It follows that \( I_{n+1} / I_n \leq I_{n+1} / I_n \leq I_{n+1} / I_n \) if \( k_n \geq k_{n+1} \) and \( I_{n+1} / I_n \leq I_{n+1} / I_n \) if \( k_n \geq k_{n+1} \). In particular, the background intensity in a layer with smaller permittivity is always larger than in an adjacent layer with higher permittivity.

An upper bound for the background intensities in each layer can now be obtained as follows: let us consider a scattering process with reflection coefficient \( R = 1 - T \). Due to the boundary conditions mentioned above (no incoming wave from the right-hand side), we have \( |A_{N+1}| = \sqrt{T} \) and \( B_{N+1} = 0 \), hence \( I_{N+1} = T = 1 - R \). The amplitudes \( |A_n| \) and \( |B_n| \) inside the rightmost layer then follow from (7) and (8), with \( k_N = \sqrt{\epsilon_{\text{high}}} \), and \( k_{N+1} = 1 \) and resulting background intensity:

\[
I_N = \frac{1 - R}{2} \left(1 + \frac{1}{\epsilon_{\text{high}}} \right).
\]

The background intensity \( I_{N-1} \) in the adjacent layer with permittivity \( \epsilon_{\text{low}} \) fulfills \( I_{N-1} \leq \alpha I_N \), where

\[
\alpha = \frac{\epsilon_{\text{high}}}{\epsilon_{\text{low}}} > 1
\]

defines the largest eigenvalue \( \lambda_n^{(2)} \) of \( M_{N-2} \cdot M_{N-2} \) (with \( k_N = \sqrt{\epsilon_{\text{high}}} \) and \( k_{N-1} = \sqrt{\epsilon_{\text{low}}} \)). In contrast, the eigenvalues of the matrix \( M_{N-2} \cdot M_{N-2} \) corresponding to the following layer (with \( k_N = \sqrt{\epsilon_{\text{low}}} \) and \( k_{N-1} = \sqrt{\epsilon_{\text{high}}} \)) are given by \( \lambda_n^{(1)} = 1 \) and \( \lambda_n^{(2)} = 1 / \alpha < 1 \), such that \( I_{N-2} \leq I_{N-1} \). For each double layer, the background intensity thereby increases at most by a factor \( \alpha \). In total, we obtain \( I_n \leq I_n^{(\text{max},r)} \), with upper bound

\[
I_n^{(\text{max},r)} = \alpha^{\left\lfloor \frac{n}{2} \right\rfloor} I_N,
\]

where \( \left\lfloor . \right\rfloor \) denotes the floor function, i.e. \( \left\lfloor n/2 \right\rfloor = n/2 \) if \( n \) is even, and \( \left\lfloor n/2 \right\rfloor = (n - 1)/2 \) if \( n \) is odd.

A similar bound can also be derived starting from the left hand side: \( A_0 = 1 \) and \( B_0 = e^{i\theta} \sqrt{R} \). Again, \( A_1 \) and \( B_1 \) follow from (7) and (8), and the maximisation over the phase \( \phi \) yields:

\[
I_n^{(\text{max})} = \frac{(1 - \sqrt{R})^2 + \epsilon_{\text{high}}(1 + \sqrt{R})^2}{2 \epsilon_{\text{high}}}.
\]

Taking once again into account the maximum amplification factor \( \alpha \) for each double layer, we obtain another upper bound for \( I_n \), i.e. \( I_n \leq I_n^{(\text{max},l)} \) with

\[
I_n^{(\text{max},l)} = \alpha I_n^{(\text{max},r)}.
\]

Thus, we obtain two upper bounds, one exponentially increasing from right to left, and one exponentially increasing from left to right. The maximal reachable background intensity within the \( n \)th layer consequently is

\[
I_n^{(\text{max})} = \min(I_n^{(\text{max},l)}, I_n^{(\text{max},r)}).
\]

The largest intensity

\[
I(\text{max}) = \max I_n^{(\text{max})}
\]

occurs in the layer \( n = n_{\text{max}} \) where the two exponential functions intersect each other. Furthermore, we choose the optimal value of the reflection coefficient \( R \) which maximises (23). For \( \epsilon_{\text{high}} = 2.3^2 \) and \( \epsilon_{\text{low}} = 1.49^2 \), we numerically find the optimal values \( R = 0.220 \) (with \( n_{\text{max}} = (N - 1)/2 \) if \( (N - 1)/2 \) is even, and \( R = 0.549 \) (with \( n_{\text{max}} = (N - 3)/2 \) if \( (N - 1)/2 \) is odd.

The above upper bound will be realised only under the condition that the amplitudes \( v_n = (A_n, B_n)^T \) inside each layer coincide with the eigenvector \( w_n^{(1)} \) or \( w_n^{(2)} \) corresponding to the required eigenvalue \( \lambda_n^{(1)} \) or \( \lambda_n^{(2)} \). According to (16), both eigenvectors have the property \( \|A_n\| = \|B_n\| \). Due to flux conservation, see (9), this property cannot be fulfilled precisely (since \( T = 0 \) implies \( A_n = B_n = 0 \), for all \( n \)), but asymptotically for large \( n \). Moreover, the requirement that, in this asymptotic limit, the transfer matrix \( M_n \) must map the vector \( w_n^{(1)} \) onto the vector \( w_n^{(2)} \) or \( w_n^{(2)} \) onto \( w_n^{(1)} \), depending on whether \( n \) is even or odd, in order to saturate the upper bound, can be used to determine the thickness of the \( n \)th layer as \( d_n = \lambda_n / 4 \) (for \( N \to \infty \)). For finite \( N \), the actual, numerically determined optimal thicknesses (see table A1 below) may differ from this asymptotic value, especially close to the sample boundaries (i.e. for \( n \approx 1 \) and \( n \approx N \), respectively), where the intensities are still small, and close to the sample centre, where the transition from exponentially increasing to exponentially decreasing intensities occurs.

In figure 2, we confirm these considerations by a comparison of the upper bound (21) and (19) with numerically optimised solutions of the wave equation (4). We see that the numerical solutions well reproduce the behaviour predicted by the theoretical upper bound. The exponential behaviour of the upper bound (23) also explains why the numerical results are rather insensitive to the precise choice of the target function (10) of the optimisation procedure, see section 2.2. Indeed, remember that the upper bound was derived by maximising the background intensity \( I_n \), as given by (13), close to the centre of the slab, whereas the numerical
optimisation maximises \( \gamma \) as defined by equation (10). Obviously, in both cases, the best strategy is to realise, as closely as possible, the maximum rate of exponential increase given by the upper bound (23)—although different choices of the target function may affect the optimal value of \( R \), which determines the intensities in the outermost layers. For example, in figure 2(b), the reflection coefficient \( R = 0.330 \) of the stack which maximises \( \gamma \) is smaller than \( R = 0.549 \) for the stack which maximises \( I_n \), since the point \( n = n_{\text{max}} \) where \( I_n \) reaches its maximum is not exactly in the centre of the stack, but slightly shifted to the left-hand side.

Nevertheless, the exponential dependence of the upper bound (23) for the background intensity suggests a similar behaviour for the optimised value \( \gamma^{(\text{max})}(N) \) of the target function \( \gamma \):

\[
\gamma^{(\text{max})}(N) \approx \sqrt{\pi} \gamma^{(\text{max})}(N - 2).
\]  

Although (24) is useful as an approximate analytical benchmark, we do not expect it to hold exactly, since \( \gamma \) depends on details of the structure—in particular the thicknesses \( d_n \) and the positions of the full intensity profile’s minima and maxima with respect to the layers containing the upconverter material—which are difficult to evaluate analytically.

3.2. Properties of optimised structures

As already mentioned in the introduction, also Bragg stacks, i.e. photonic structures which consist of layers with periodically alternating thicknesses \( d_i = d / (4 \sqrt{\varepsilon_i}) \) (which are mainly known for their ability to achieve high reflectivity at a certain wavelength regime [10]), lead to an enhanced upconversion efficiency [7]. There, the only optimisation parameter is the design wavelength \( d \) [7]. It is therefore to be expected that our present approach with the individual layer thicknesses \( d_n \) as tunable control parameters enables further improvement, as we demonstrate hereafter.

3.2.1. Local intensity profile

To start with, figure 3 shows the intensity profiles \( I(z) \) for a photonic structure with individually optimised thicknesses (a), and for the optimal Bragg stack (b). As an example of experimentally realistic parameters [7], we choose \( \omega_0 = \omega_{\gamma + 1} = 1 \), \( \omega_{\text{high}} = 2.3^2 \) and \( \omega_{\text{low}} = 1.49^2 \) for the permittivities of the respective materials, for a structure with \( N = 31 \) layers. In figure 3(a), we see that, as a consequence of the individually optimised layer thicknesses—with the upconverting layers’ thicknesses increasing from the sample edge to its centre (for specific values, see table A1 in appendix), the maximum intensity reached within the structure is more than 1000 times larger than the incident intensity. In the case (b) of the Bragg stack, the intensity is also enhanced, but by less than a factor 20, with an optimised design wavelength \( d = 1.149\lambda \), where \( \lambda = 2\pi c / \omega \) denotes the wavelength of the incident wave in vacuum. In both, (a) and (b), \( I(z) \) has its maxima within the layers with low permittivity (which contain the upconverter material) and, correspondingly, minima in the layers with higher permittivity. This is expected as a consequence of optimising the intensity enhancement factor \( \gamma \) inside those layers, which contain the upconverter, see (10). This factor results as \( \gamma \approx 220.4 \) and \( \gamma \approx 12.8 \) in (a) and (b), respectively.

3.2.2. Enhancement of intensity for increasing number of layers

Let us now examine the scaling behaviour of the intensity enhancement factor \( \gamma \) with the number \( N \) of layers, see figure 4. For large \( N \), the structure with individually optimised thicknesses (solid line) exhibits an exponential increase, which approximately agrees with the analytical
benchmark (24) (grey dashed–dotted line). But even for small samples with only \( N = 11 \) layers, individual optimisation still leads to an improvement by a factor of approximately 1.8 with respect to the Bragg stack (dashed lines).

Furthermore, figure 4 also quantifies the impact of uncontrolled deviations from the optimised structures as unavoidably induced by fabrication errors. We allow for statistical variations of the \( d_n \), which we sample from a Gaussian distribution with standard deviation \( \sigma \), chosen as a certain fraction (1\% or 3\%) of the desired mean value \( d_{n_0} \), and determine the intensity enhancement factor \( \gamma \) averaged over 10,000 random configurations (symbols). We see that the structure with individually optimised thicknesses (filled symbols) is more sensitive against errors than the Bragg stack (open symbols). This is expected, since, by construction, any deviation from the fully optimised structure decreases \( \gamma \), whereas, in case of the Bragg stack, deviations may also increase \( \gamma \), even if reducing it on average. For the fully optimised structure subject to random errors, the enhancement factor starts to deviate from the exponentially increasing dependence on \( N \), as predicted by (24) at a certain number of layers (which depends on \( \sigma \)), and no longer increases (or even slightly decreases) for larger \( N \). However, the fully optimised design retains its advantage over the Bragg stack even under the influence of errors for small layer numbers \( N \). For instance the enhancement factor of the optimised design with \( N = 15 \) and errors of size 3\% is 1.66 times larger than for a Bragg stack with the same distribution of fabrication errors and number of layers.

3.3. Robust optimised structures

The sensitivity of our structures against fabrication errors raises the question whether it is possible to modify our structures such as to render them more robust with respect to these errors. A direct optimisation of the average enhancement factor \( \gamma \) as defined above, however, is not feasible due to the large number of realisations needed to determine the average. Instead, we will consider in this section random variations of the wavelength \( \lambda \) of the incident light. With view at the upconversion scenario described in the introduction, these must be taken into account in order to achieve efficient upconversion not only under illumination with a monochromatic laser, but also for incoherent incident light exhibiting a finite spectral bandwidth. In this case, the relevant spectral window is determined by the absorption spectrum of the upconverter ions. For the case of \( \beta \)-NaErF\(_{4}\)\(\_\)Y\(_{0.2}\)F\(_{3}\), the same upconverter erbium ions as in [7], the relative width \( \Delta \lambda/\lambda_0 \) of this spectrum around its maximum at \( \lambda_0 \approx 1523 \) nm roughly equals \( \sigma = 2.5\% \) [21]. We will therefore consider

\[
\gamma = \frac{1}{2\sigma} \int_{-\infty}^{+\infty} ds \, \gamma((1+s)\lambda_0), \tag{25}
\]

as our new optimisation target, where \( \gamma(\lambda) \) is the intensity enhancement factor as defined by equation (10) with wavelength \( \lambda \) of the incident light. We approximate the integral in equation (25)—where, for the purpose of an efficient numerical evaluation, the absorption spectrum is modelled by a rectangular distribution—by a discrete sum over \( \approx 50 \) equally spaced points in the interval \( s \in [-\sigma, \sigma] \). This increases the time needed for the optimisation by a factor 50, which is still feasible. We have checked that, up to \( N \approx 21 \), the function \( \gamma(\lambda) \) for the optimised structures remains sufficiently
smooth such that the discrete sum can be applied with good approximation.

Since the only relevant parameters of our system are the layer thicknesses in units of the incident wavelength $\lambda$, a change of $\lambda$ has the same effect as a multiplication of all layer thicknesses by a constant factor. Intuitively, we expect structures which are more robust with respect to a systematic (i.e. constant for all layers) error of the thicknesses to be more robust also against statistical errors. This expectation is confirmed by figure 5, where we plot $\gamma$ without statistical errors (black filled symbols) and with statistical errors of relative size 3% (grey filled symbols) and 10% (open symbols) for three different types of structures: (a) those discussed in section 3.2 which maximise $\gamma$; (b) our new robust optimal structures which maximise $\gamma$, and (c) the optimal Bragg stack (which, as we have checked, is almost the same when optimised with respect to $\gamma$ or $\tau$). In (a), we see that the spectrally averaged $\gamma$ as defined by equation (25) without statistical errors (black filled symbols) is strongly reduced for large $N$ as compared to $\gamma$ shown in figure 4. For these structures, $\gamma(\lambda)$ exhibits an extremely narrow peak due to the formation of a strongly localised and narrow eigenmode. Consequently, these structures are also sensitive against statistical errors, see figure 5(a). With our new robust structures, figure 5(b), it is possible to increase the spectrally averaged $\gamma$ up to $\gamma \approx 5.2$ (without statistical errors) and up to $\gamma \approx 2.8$ ($\gamma \approx 4.9$) in the presence of statistical fabrication errors as large as 10% (3%). This is approximately 1.5 (1.8) times larger than for a Bragg stack with the same statistical error, see figure 5(c).

Finally, we note that, for weak irradiation, the upconversion efficiency is expected to scale quadratically with the intensity, see the discussion after equation (10). If we account for that by replacing $R(z)$ by $I^2(z)$ in the definition of $\gamma$, the advantage of our structures is even more pronounced: for example, for $N = 15$, we obtain $\gamma_{\text{rob}} \approx 27.8 (\gamma_{\text{rob}} \approx 57.4$) for the robust optimised structures with 10% (3%) errors, as compared to $\gamma_{\text{rob}} \approx 8.9 (\gamma_{\text{rob}} \approx 12.4$) for the Bragg stack.

4. Conclusion

We have shown how the individual tuning of the layer thicknesses of a two-component photonic structure allows to push the irradiance concentration in the sample’s upconverting sub-volume close to an analytically derived upper bound.

The physical mechanism responsible for this enhancement is the appropriate fine-tuning of interferences between wave amplitudes which are multiply reflected within the multilayer stack. This makes it possible to trap photons of a given wavelength inside the stack, for a time which increases exponentially with the number of layers. The rate of the exponential increase is proportional to the ratio $\alpha = \epsilon_{\text{high}}/\epsilon_{\text{low}}$ of the permittivities of the two materials forming the multilayer stack (or, equivalently, to the square of the ratio of their refractive indices). Random variations around the optimal structure limit this increase, but the optimised structure still retains its advantage over periodic stacks. Further accounting for the effects of a finite spectral bandwidth of the incoherent incident light, we find robust optimised structures more suitable to achieve efficient upconversion within a realistic spectral window. While these do not exhibit the exponential increase in integrated intensity with additional layers, they still perform significantly better than the Bragg stack, for not too large numbers of layers and even in the presence of fabrication errors as large as 10%.

We thus conclude that the optimised design of the multilayer structure proposed in the present paper bears a considerable potential for applications, such as upconversion, which benefit from an enhancement of the local field intensity.
Table A1. Table with normalised layer thicknesses $d_n$ for optimised structures and their robust counterparts of variable layer number $N$. Thicknesses are given in units of $\lambda_n/4 = \lambda/(4\sqrt{\varepsilon_n})$, $n = 1, \ldots, N$, where $\lambda$ denotes the wavelength of the incident light in vacuum. The permittivities of the layers are $\varepsilon_n = \varepsilon_{\text{high}}$ for odd $n$, and $\varepsilon_n = \varepsilon_{\text{low}}$ for even $n$, with $\varepsilon_{\text{high}} = 2.3^2$ and $\varepsilon_{\text{low}} = 1.49^2$.

| Layer $n$ | $N = 9$ | $N = 11$ | $N = 19$ | $N = 21$ | $N = 9$ | $N = 11$ | $N = 19$ | $N = 21$ |
|----------|---------|---------|---------|---------|---------|---------|---------|---------|
| 1        | 1.11398 | 1.06867 | 1.03937 | 1.04823 | 1.19128 | 1.14659 | 1.21284 | 1.21400 |
| 2        | 1.01141 | 0.99331 | 0.93387 | 0.90776 | 1.14891 | 1.05583 | 0.85162 | 0.84232 |
| 3        | 1.12345 | 1.06185 | 1.06688 | 1.08878 | 1.15104 | 1.08278 | 1.19416 | 1.19979 |
| 4        | 1.80169 | 1.67730 | 0.96246 | 0.92541 | 1.68636 | 1.75104 | 1.30070 | 1.27379 |
| 5        | 1.09146 | 1.15063 | 1.05095 | 1.07920 | 1.11107 | 1.08370 | 0.94262 | 0.94974 |
| 6        | 0.99517 | 1.26261 | 1.05378 | 0.97555 | 0.96507 | 1.07297 | 1.71273 | 1.71396 |
| 7        | 1.13429 | 1.14682 | 1.00226 | 1.05291 | 1.18483 | 1.22258 | 0.98444 | 0.96121 |
| 8        | 0.88347 | 0.89420 | 1.78197 | 1.19220 | 0.82686 | 0.79341 | 1.20109 | 1.25001 |
| 9        | 1.08205 | 1.17044 | 0.96158 | 0.95619 | 1.11575 | 1.28359 | 1.21887 | 1.20612 |
| 10       | 0.83459 | 1.38801 | 1.88968 | 0.70521 | 0.78642 | 0.80461 |
| 11       | 1.09510 | 1.05927 | 0.97692 | 1.16234 | 1.37103 | 1.36919 |
| 12       | 0.98197 | 1.09973 | 0.58789 | 0.58810 |
| 13       | 1.09218 | 1.04206 | 1.47898 | 1.48860 |
| 14       | 0.91516 | 0.97629 | 0.47013 | 0.45626 |
| 15       | 1.10380 | 1.06189 | 1.55946 | 1.58204 |
| 16       | 0.89289 | 0.94071 | 0.39042 | 0.36247 |
| 17       | 1.10836 | 1.06937 | 1.62297 | 1.66100 |
| 18       | 0.88445 | 0.92756 | 0.32991 | 0.28573 |
| 19       | 1.05394 | 1.07235 | 1.33218 | 1.73610 |
| 20       | 0.92243 | 0.21127 |
| 21       | 1.03724 | 1.39337 |

(or, equivalently, the local irradiance) due to more efficient trapping of the incident photons. While our present paper focuses on the enhancement of the intensity, there are several additional aspects that need to be addressed in future work: indeed, the upconversion luminescence does not only depend on the local irradiance, but, e.g., also on the photonic density of states (which controls the spontaneous emission rates between the atomic energy levels of the upconverter), and on the efficiency of energy transfer processes relevant for upconversion. The resulting upconversion luminescence can be estimated using a rate equation model [6]. Moreover, absorption of the incident photons by the upconverter material must be taken into account in a more complete description, with the aim of achieving a reliable prediction and optimisation of the full upconversion efficiency in experimentally realisable photonic structures, under realistic manufacturing constraints.

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Appendix. List of optimised thicknesses

Table A1, lists exemplary structure information on optimised structures (left) and their robust counterparts (right), for $N = 9, 11, 19$ and $N = 21$ and materials $\varepsilon_{\text{high}} = 2.3^2$ and $\varepsilon_{\text{low}} = 1.49^2$. Note that most layer thicknesses $d_n$ are close to $1$ (in units of $\lambda_n/4$). For the optimal structures (left), only layers with even $n$, close to the centre of the stack (i.e., $n = 4$ for $N = 9$ and $N = 11$, $n = 8$ for $N = 19$, and $n = 8$ and $10$ for $N = 21$) are exceptionally thick. As we have checked, these are the layers where the highest intensities are achieved. The structures optimised for a finite spectral bandwidth are very similar to the non-robust optimised multilayer stacks for small $N$. For increasing $N > 17$, however, additional layers can not increase the intensity over the full spectral bandwidth any more, see figure 5(b). Comparing $N = 19$ with $N = 21$ (right), we see that layers near the incident light are almost unchanged and thin layers with $\varepsilon_{\text{low}}$ (even $n$) on the rear side are added having only little influence on the intensity profile in the relevant wavelength regime.
ORCID iDs

Fabian Spallek https://orcid.org/0000-0002-3103-0275

References

[1] Brown G and Wu J 2009 Laser Photon. Rev. 3 394–405
[2] Sark W G v, Wild J d, Rath J K, Meijerink A and Schropp R E 2013 Nanoscale Res. Lett. 8 81
[3] Trupke T, Green M A and Würfel P 2002 J. Appl. Phys. 92 1668–74
[4] Richards B S 2006 Sol. Energy Mater. Sol. Cells 90 2329–37
[5] Fischer S, Favilla E, Tonelli M and Goldschmidt J C 2015 Sol. Energy Mater. Sol. Cells 136 127–34
[6] Herter B, Wolf S, Fischer S, Gutmann J, Bläsi B and Goldschmidt J C 2013 Opt. Express 21 A883–900
[7] Hofmann C L M, Herter B, Fischer S, Gutmann J and Goldschmidt J C 2016 Opt. Express 24 14895
[8] Pollnau M, Gambelin D R, Lüthi S R, Güdel H U and Hehlen M P 2000 Phys. Rev. B 61 3337–46
[9] Agarwal G S 1975 Phys. Rev. A 11 253–64
[10] Yeh P and Yariv A 1976 Opt. Commun. 19 427–30
[11] Redfield D 1974 Appl. Phys. Lett. 25 647–8
[12] Yablonovitch E 1982 J. Opt. Soc. Am. 72 899–907
[13] Vynck K, Burresi M, Riboli F and Wiersma D S 2012 Nat. Mater. 11 1017–22
[14] Scholak T, de Melo F, Wellens T, Mintert F and Buchleitner A 2011 Phys. Rev. E 83 021912
[15] Scholak T, Mintert F, Wellens T and Buchleitner A 2010 Transport and entanglement semiconductors and semimetals Quantum Efficiency in Complex Systems, Part I: Biomolecular systems ed E R Weber et al vol 83 (Amsterdam: Elsevier) ch 1, pp 1–38
[16] Walschaers M, Schlawin F, Wellens T and Buchleitner A 2016 Annu. Rev. Condens. Matter Phys. 7 223–48
[17] Walschaers M, Mulet R and Buchleitner A 2017 J. Phys. B: At. Mol. Opt. Phys. (arXiv:1704.07168 [cond-mat, physics: quant-ph]) submitted
[18] Abélès F 1950 Ann. Phys., Paris 5 596
[19] Press W H, Teukolsky S A, Vetterling W T and Flannery B P 2007 Numerical Recipes: The Art of Scientific Computing 3rd edn (New York: Cambridge University Press) http://numericalrecipes/
[20] Nelder J A and Mead R 1965 Comput. J. 7 308–13
[21] Fischer S, Steinkemper H, Löper P, Hermle M and Goldschmidt J C 2012 J. Appl. Phys. 111 013109