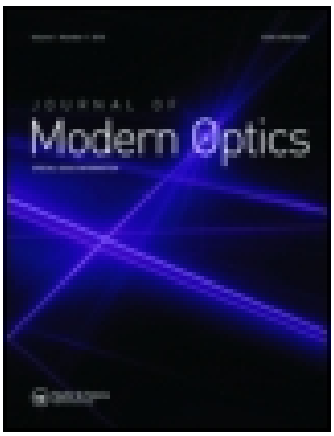


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# Enhanced third-order nonlinear optical response of photonic bandgap materials

Robert L. Nelson [a b] & Robert W. Boyd [b]

[a] Materials and Manufacturing Directorate, Air Force Research Laboratory , (MLPO), Wright-Patterson Air Force Base, Ohio, 45433-7707, USA

[b] Institute of Optics, University of Rochester , Rochester, New York, 14627, USA

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# Enhanced third-order nonlinear optical response of photonic bandgap materials

ROBERT L. NELSON†‡ and ROBERT W. BOYD‡

† Materials and Manufacturing Directorate, Air Force Research Laboratory, (MLPO), Wright–Patterson Air Force Base, Ohio 45433-7707, USA

‡ Institute of Optics, University of Rochester, Rochester, New York 14627, USA

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**Abstract.** We calculate the nonlinear phase shift acquired by a laser beam in propagating through a one-dimensional photonic bandgap material, that is a material in which the linear refractive index is periodically modulated along the direction of propagation. We find that the nonlinear phase shift shows resonances for laser frequencies close to the edge of the stop band of the photonic bandgap structure. Enhancements of the nonlinear phase shift compared with that of a homogeneous nonlinear optical material by a factor of approximately five are predicted under realistic laboratory conditions. We find that similar enhancements of the two-photon absorption rate can occur for a material with an imaginary nonlinear susceptibility. We also treat the case of a photonic bandgap material containing a 'defect,' that is a central region somewhat too thick to conform to the periodicity of the system, and find that the nonlinear phase shift can be enhanced by a factor of approximately thirty.



## 1. Introduction

A photonic bandgap (PBG) structure is a material system that prohibits the propagation of electromagnetic radiation over some range of wave-vectors. Incident radiation with these wave-vectors will be almost entirely reflected because of the interference properties of radiation within the structure. PBG structures can be constructed in one, two and three dimensions. Here we consider only the one-dimensional case as it is the simplest to examine and construct and yet still is potentially useful. The principles involved in one-dimensional structures can be extended to higher dimensions, however.

The properties of a PBG material can be engineered by selecting proper materials for the constituents and by altering the mechanical structure. A widely analysed one-dimensional structure is a periodic multilayer of two materials possessing different linear optical properties and whose optical layer thicknesses are near a quarter-wavelength [1]. Interesting nonlinear effects can occur in these PBG structures as will be discussed below. Others have studied spontaneous emission rates [2–5] and enhanced gain in a PBG laser [6]. Higher dimensional arrays of spheres or cylinders have been shown to have PBG behaviour [2, 7, 8].

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Enhanced nonlinear properties of periodic dielectric structures were predicted as early as 1970 by Bloembergen and Sievers [9], and later by Tang and Bey [10], who introduced the idea of harmonic phase matching by using the optical properties of multilayers. Second-harmonic generation (SHG) has been investigated by a number of workers in various types of multilayer structure. Enhancement of the reflected second harmonic was observed experimentally in a 17-layer-pair structure [11] where the fundamental was tuned to the middle of the PBG stop band providing for strong counterpropagating beams. Enhanced SHG using a defect in an otherwise periodic structure has been investigated theoretically and experimentally [12, 13] and was also experimentally demonstrated in a vertical cavity geometry of GaAs/AlAs multilayers combined with a $SiO_2/TiO_2$ distributed Bragg reflector [14]. Enhancement of SHG in fibre Bragg gratings was also the subject of recent work [16].

Enhanced SHG in a one-dimensional PBG structure without accompanying cavity modes was demonstrated numerically for large-index modulated structures with a pulsed incident fundamental [17] and theoretically for weakly periodic media using multiple-scales perturbation [18]. In these references, enhancements, over an equivalent length of phase matched bulk material, of two to three orders of magnitude in power levels of the generated second harmonic when the fundamental was tuned near the photonic band edge were reported. These results provide strong motivation for further investigation and application of such structures. There are three reasons for the unusually strong nonlinear optical response. First the field amplitudes are enhanced owing to resonance effects, second the transmission factor is large so that most of the fundamental energy is transmitted (also in [17] the second harmonic is tuned near the second-order band edge), and third the group velocity at the band edge is small so that the fundamental field spends more time inside the structure which provides greater conversion efficiency.

Third-order processes have also gained attention in PBG-type materials with investigations into gap–soliton propagation and optical switching [19–29]. An interesting application involving a $\chi^{(3)}$ process was the nonlinear optical diode in [30, 31] where optical transmission was dependent on the direction of propagation. Recently three different one-dimensional PBG structures were investigated for their optical limiting abilities [32], and in the investigation of a quarter-wavelength type structure, good broad-band limiting properties of a 63-layer structure with modest linear index modulation but nonlinear coefficients of equal value but opposite sign in the adjacent layers were numerically found. The limiting process was accomplished through strong reflectivity at high incident intensities.

In the present article we numerically further investigate the nonlinear optical properties of PBG structures with an emphasis on the question of how the nonlinear response compares with the bulk response of the nonlinear constituent. Unlike in [17, 32] we shall consider systems where only one of the materials possesses the dominant nonlinear response. Also we shall concentrate on deep index modulations, with relatively weak nonlinearities, where properties such as reflection and transmission are relatively unaffected by the nonlinear interactions occurring inside the structure, unlike [32].

## 2. Method of calculation

We consider plane waves of linear polarization propagating in the direction perpendicular to the plane of the layers in the multilayered PBG structure. The structures considered are composed of $N$ layer pairs with only one of the constituents having a third-order Kerr optical nonlinearity. Initially we consider the material with only the linear response as constituent a and the material with the third-order nonlinearity is denoted constituent b. The refractive index for the b layers is given by

$$n_b = n_{b0} + 2\bar{n}_2|\mathbf{E}(\omega)|^2, \tag{1}$$

where $\bar{n}_2$ is the nonlinear refractive index. Initially the zeroth-order field magnitudes throughout the structure are calculated using the incident field magnitude and assuming that no nonlinearities present. Also we shall suppress the explicit $\omega$ notation as it is understood that all field quantities are oscillating at the same optical frequency.

The field solutions are arrived at by requiring that the electromagnetic boundary conditions of continuity of the tangential components of $E$ and $H$, with $E$ and $H$ plane wave solutions to Maxwell's equations in each layer, be satisfied simultaneously at each of the $2N + 1$ boundaries. The incident $E$ and $H$ fields are considered to have a fixed amplitude. The unknown $H$ fields are expressed in terms of the $E$ fields using the plane-wave relation

$$\mathbf{H} = n\hat{\mathbf{k}} \times \mathbf{E}. \tag{2}$$

For the zeroth-order solution this leads to the matrix equation

$$\mathbf{AF} = \mathbf{F}_0, \tag{3}$$

where $\mathbf{A}$ is a $(4N + 2) \times (4N + 2)$ matrix representing the two boundary conditions at each interface, $\mathbf{F}$ is a vector representing all the unknown electric field amplitudes including the backward-travelling amplitudes (two per $2N$ layers plus reflected and transmitted; $4N + 2$ in all) and $\mathbf{F}_0$ is the vector that represents the known input amplitudes which for this case is just the incident $E$ and $H$ fields. This approach is equivalent to the transfer matrix technique and details may be found elsewhere [1].

The field amplitudes from the zeroth order solution are used to calculate the first-order correction to the refractive indices in the nonlinear layers. Considering the field in layer $j$ in more detail, we write

$$\mathbf{E}_{0j} = \hat{\mathbf{e}}(E_{0j}\,e^{ikz} + E'_{0j}\,e^{-ik'z}), \tag{4}$$

where a prime denotes a backward-travelling wave quantity, $k = n\omega/c$ and $k' = n'\omega/c$. The refractive indices in the expressions for the wavenumbers include both the linear and the nonlinear contributions and, as we explain below, are generally different for the forward- and backward-moving waves. The scalar amplitudes are those at the top of layer $j$ and the exponentials describe the spatial variation within the layer. When proper account is taken of the spatial dependences of all the terms in the full expression for $|\mathbf{E}_{0j}|^2$, it is noted that only the terms which contain the same spatial dependence of the moving wave should be retained [33]. Therefore, for the forward moving wave, we have

$$n_{bj} = n_{b0} + 2\bar{n}_2(|E_{0j}|^2 + 2|E'_{0j}|^2) \tag{5}$$

and for the backward-travelling wave

$$n'_{bj} = n_{b0} + 2\bar{n}_2(2|E_{0j}|^2 + |E'_{0j}|^2) \tag{6}$$

as the expressions for the refractive indices. The differing indices for the two waves arise out of the unequal strengths of self- and cross-phase modulation as has been described in [33, 34]. Previous work [32] does not seem to have maintained the distinction. Also we use the slowly varying amplitude approximation as we are not necessarily interested in the strong nonlinear limit. Once the new $n_{bj}$ and $n'_{bj}$ are calculated, another self-consistent solution is achieved by a second iteration of equation (3) using the new values of the layer indices separately for the forward- and backward-travelling waves. This first order solution for the fields is usually accurate enough in the weakly nonlinear limit (either weak field or weak $n_2$) to be used as a representation for the nonlinear material response. In some situations, however, we may gain accuracy by continuing the iteration process until a convergence to within specified limits is achieved. This can be noticeable at higher field strengths or nonlinearities. At higher field strengths or nonlinearities, the solution begins an oscillatory behaviour with the number of iterations; that is it does not converge to a single solution. At lower nonlinear interaction levels the solution asymptotically converges to a steady value. When there is asymptotic behaviour in the solution, then it is assumed that the answer provided for the nonlinear response is satisfactory while oscillatory solutions are avoided. The oscillatory solution may indicate an underlying dynamical instability, but we have not yet verified that hypothesis.

### 3.1. *Non-dissipative photonic bandgap structures*

By using the procedure described above, the field magnitudes in all layers including the exiting field can be determined. The algorithm provides amplitudes and phases for all fields and allows consideration of the nonlinear phase shift on passage of the incident wave through a nonlinear medium. By comparing the nonlinear phase shift between PBG structures and homogeneous media, the relative effective nonlinearity of the PBG can be determined. Of particular interest is whether the nonlinear phase shift is larger on passage through a PBG material than on passage through a homogeneous film of equivalent total thickness.

All calculations are performed assuming a fixed incident wavelength of 1.06 μm and all layers are assumed to be lossless dielectrics. The surrounding medium has an index of 1.0 and initially the indices of the two constituents are set at $n_a = 1.5$ and $n_b = 2.0$ with material b being the Kerr active medium. There are ten layer pairs and as a first example the thickness of material a (the low-index linear medium) is varied as the independent variable. The index in the nonlinear active layer is described as

$$\begin{aligned} n_{bj} &= n_{b0} + 2\bar{n}_2(|E_{0j}|^2 + 2|E'_{0j}|^2), \\ n'_{bj} &= n_{b0} + 2\bar{n}_2(2|E_{0j}|^2 + |E'_{0j}|^2), \end{aligned} \tag{7}$$

where in performing this calculation we have assumed that the intensity of the incident light was adjusted so that $2\bar{n}_2|E_0|^2 = 10^{-6}$.

Figure 1 displays three curves; the nonlinear phase shift through the PBG structure, the nonlinear phase shift through a homogeneous film constructed with

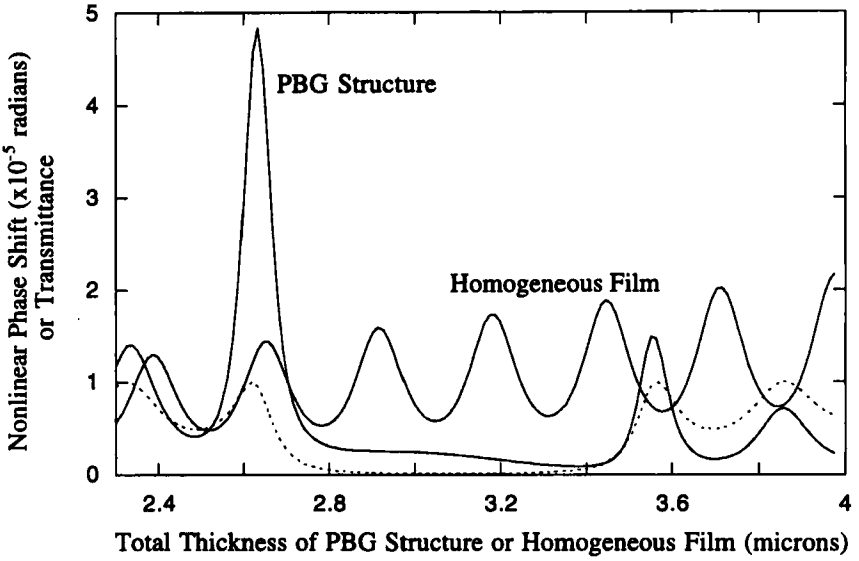


Figure 1. Nonlinear phase shift produced by a PBG structure as compared with that of a homogeneous film of the nonlinear constituent with the same total thickness as the PBG structure. The broken curve is a plot of the transmittance used to locate the position of the band edge. The thickness of the high-index nonlinear component is set at the quarter-wave thickness and the thickness of the low-index layer is allowed to vary in the PBG structure.

material b of the same total thickness as the PBG structure, and a plot of the transmittance of the PBG structure to display qualitatively where the band edge and bandgap are located. As might be inferred from the results of [17], there is an enhancement of the nonlinear phase shift at the band edge of the PBG material over the response of the homogeneous layer. Since forward degenerate four-wave mixing (DFWM) is an automatically phase-matched process, some features present in harmonic generation do not appear. Together with phase matching, there is no 'extra' enhancement by tuning the harmonic to a higher-order band edge; therefore the maximum achievable enhancement with DFWM is probably smaller. Certainly the high transmittance and the resonantly enhanced fields inside the high-index layers are contributing to the enhancement.

Further investigation shows that the size of the enhancement effect increases with increasing number of layer pairs and with increasing depth of the index differential. The relative-thicknesses of the individual layers can vary considerably around the quarter-wave thickness and still provide bandgap effects. This latitude in PBG design can provide for better enhancements in PBG structures. The optimum enhancement of the nonlinear phase shift, in the ten-layer-pair structure that we described above, occurred when the low-index layer was approximately 65% of its quarter-wave thickness and the nonlinear high-index layer was approximately 110% of its quarter-wave thickness. All quarter-wave thicknesses in this paper are defined with respect to the zeroth-order refractive indices of the layers.

The next example (figure 2) considers the alternative arrangement where we assume that the nonlinear constituent is the low-index dielectric layer. Now the enhancement occurs at the opposite band edge and, when optimized, the enhance-

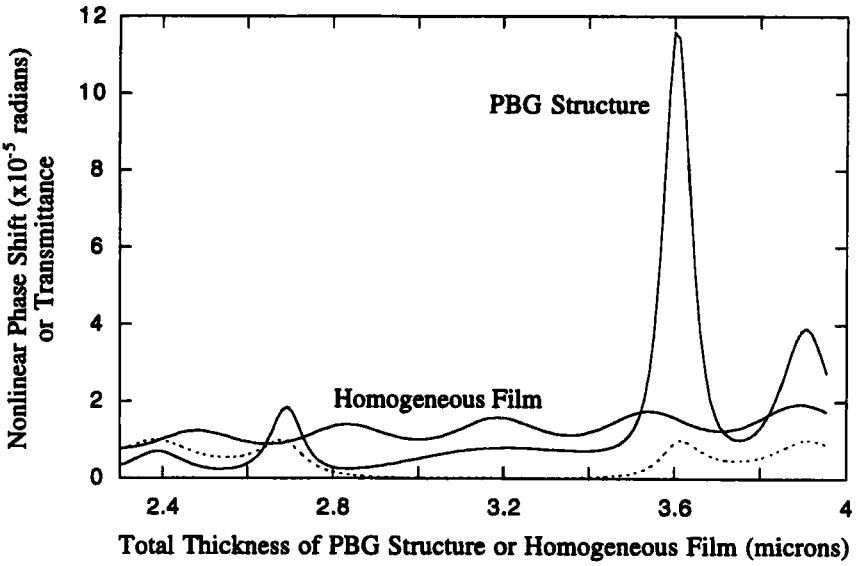


Figure 2. PBG nonlinear phase shift enhancement with the low-index constituent as the nonlinear material, optimized for maximum enhancement. The broken curve is the transmittance. The thickness of the high-index constituent is set at 85% of its quarter-wave thickness while the low-index layer thickness is allowed to vary for the PBG structure. As in figure 1, the homogeneous film has a thickness equal to the total thickness of the PBG structure and is considered to be composed of the nonlinear constituent.

ment is larger than before. Optimization is achieved by setting the high-index passive layer to 85% of its quarter-wave thickness and the low-index nonlinear layer to 140% of the quarter-wave thickness. Enhancement by a factor of approximately six over the homogeneous material is seen while a maximum enhancement factor of about four was observed when the nonlinear constituent was the high-index layer. It is clear from these results that PBG structures can provide enhancements in the nonlinear phase shift in a manner similar to the reported enhancement in SHG.

### 3.2. *Nonlinear absorption in photonic bandgap structures*

We see that band edge enhancements in lossless dielectric structures can be large and potentially useful. Alternatively there has been interest in nonlinear absorbing materials, especially in the application of optical limiting. We would expect that, since nonlinear absorption is described by an imaginary $\chi^{(3)}$, there is reason to expect similar enhancement of the nonlinear absorption. Two-photon absorption materials are promising as a means for optical limiting and are modelled as a positive imaginary contribution to the nonlinear refractive index. The action of a two-photon absorber is then to provide an intensity-dependent absorption coefficient that increases with increasing intensity. We now investigate whether a PBG-type material can also enhance the intensity-dependent absorption.

The nonlinear layers (low-index layers) have indices represented by the equations

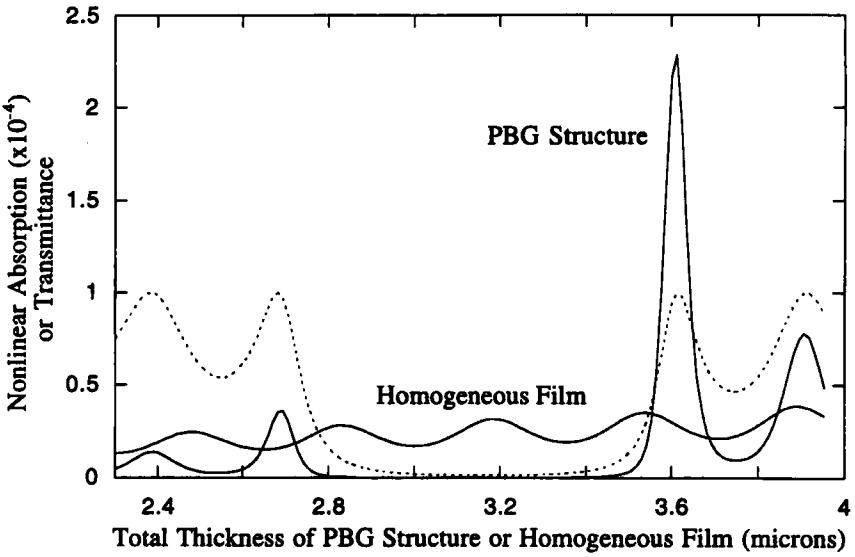


Figure 3. Change in the transmittance due to nonlinear absorption in the sample. The low-index layer is the nonlinear constituent and is allowed to vary in thickness. The high-index layer thickness is held constant at 85% of its quarter-wave thickness in the PBG structure. The thickness of the homogeneous layer is equal to the total thickness of the PBG structure. The homogeneous layer is composed of the low-index nonlinear constituent.

$$n_{aj} = n_{a0} + 2\bar{n}_2(|E_{0j}|^2 + 2|E'_{0j}|^2),$$
$$n'_{aj} = n_{a0} + 2\bar{n}_2(2|E_{0j}|^2 + |E'_{0j}|^2), \tag{8}$$

where $\bar{n}_2$ is a complex number, the imaginary part of which describes nonlinear absorption. In order to avoid the possibility that the real part of $\bar{n}_2$ (nonlinear index) could affect the magnitude of the output field, it is set to zero in order to isolate the effects of nonlinear absorption. Calculations are performed as before with several iterations done in order to ensure convergence of the solution. In this case, however, it is the magnitude of the output field and not the phase that is of interest. The linear indices of the constituent layers are assumed to be purely real, possibly modelling a non-resonant nonlinearity, and set at $n_a = 1.5$ and $n_b = 2.0$ and again there are ten layer pairs. As expected (figure 3), there is a band edge enhancement of the nonlinear absorption. The enhancement factor is about the same as in the nonlinear phase shift example.

### 3.3. *Photonic bandgap structures with a central phase slip*

There have been previous investigations into PBG structures containing a central region of thickness different from those of the other layers [11–13, 28, 29] with reported enhancements of second-harmonic generation in particular [11–13]. It is interesting to ask how the calculation that we have performed above is affected by the introduction of a central nonlinear active layer that is of different thickness from the other nonlinear layers. For consideration we take again the nonlinear constituent to be the lower refractive index layer and introduce a 0.5 μm low-index nonlinear active central layer surrounded on either side by 5 layer pairs of PBG-

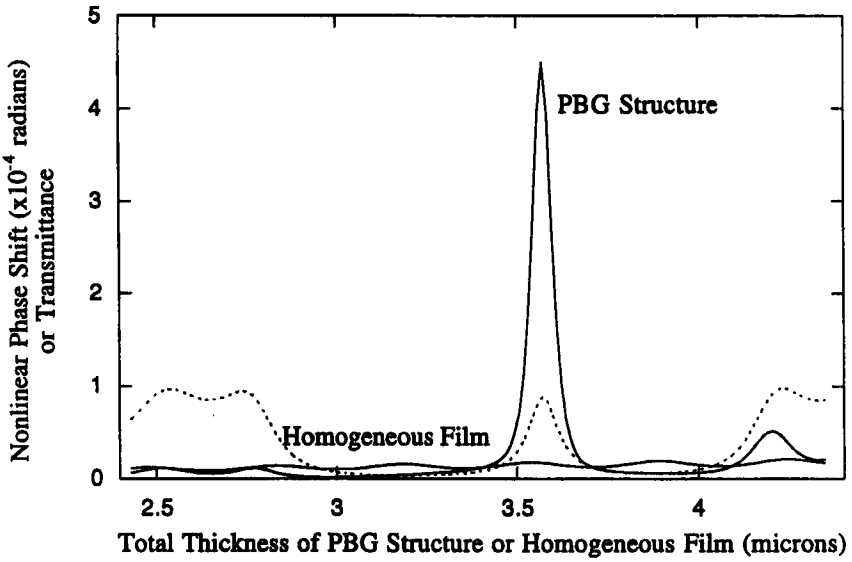


Figure 4. Response of a PBG structure with a thick central nonlinear layer. The broken curve is the transmittance. In the PBG material surrounding the central active region, the high-index layers were held fixed at 85% of their quarter-wave thickness and the low-index nonlinear layers were allowed to vary in thickness.

type material. The high index layer thickness was set at 85% of its quarter-wave thickness and, as can be seen from figure 4, maximum enhancement occurs when the low-index nonlinear material is approximately 120% of its quarter-wave thickness. Also apparent is the fact that the magnitude of the enhancement has increased to a factor of about 30 over an equivalent thickness homogeneous layer of material a. The largest enhancement occurs at the new propagation mode introduced into the bandgap. The enhancement here is analogous to the enhanced SHG reported in [12, 13] and related to the results in [29, 30]. Note that there is still an enhancement at the right band edge.

It appears that, if an attempt is made to provide maximum possible enhancement of the nonlinear response over a homogeneous material, the best situations may not be limited to strictly periodic materials. The optimization problem here is more complex since there are more degrees of freedom in the design.

## 4. Conclusions

PBG materials are potentially useful materials for the development of material systems with an enhanced nonlinear response. We have numerically demonstrated a considerable enhancement in the nonlinear refractive index at the photonic band edge over equivalent homogeneous materials. Band edge enhancements have also been shown to exist for SHG [17, 18]. Greater enhancements in the third-order nonlinearity seem possible in PBG structures with a central phase slip, or defect mode. Our results indicate an enhancement of a factor of 30, with no attempt to optimize all the physical parameters involved, in such a structure. It is likely that greater application for these structures will be found in future work.

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