

This work was written as part of one of the author's official duties as an Employee of the United States Government and is therefore a work of the United States Government. In accordance with 17 U.S.C. 105, no copyright protection is available for such works under U.S. Law. Access to this work was provided by the University of Maryland, Baltimore County (UMBC) ScholarWorks@UMBC digital repository on the Maryland Shared Open Access (MD-SOAR) platform.

Please provide feedback

Please support the ScholarWorks@UMBC repository by emailing scholarworks-group@umbc.edu and telling us what having access to this work means to you and why it's important to you. Thank you.

PROCEEDINGS OF SPIE

[SPIDigitalLibrary.org/conference-proceedings-of-spie](https://www.spiedigitallibrary.org/conference-proceedings-of-spie)

Phase matching in the presence of feedback: higher-order terms and enhancement of second-harmonic generation

Centini, Marco, D'Aguanno, Giuseppe, Sciscione, Letizia, Sibilìa, Concita, Bertolotti, Mario, et al.

Marco Centini, Giuseppe D'Aguanno, Letizia Sciscione, Concita Sibilìa, Mario Bertolotti, Michael Scalora, Mark J Bloemer, "Phase matching in the presence of feedback: higher-order terms and enhancement of second-harmonic generation," Proc. SPIE 5360, Photonic Crystal Materials and Devices II, (9 July 2004); doi: 10.1117/12.527930

SPIE.

Event: Integrated Optoelectronic Devices 2004, 2004, San Jose, CA, United States

Phase Matching in the Presence of Feedback: Higher order terms and Enhancement of Second Harmonic Generation.

Marco Centini^{*a,b}, Giuseppe D'Aguanno^{a,b}, Letizia Sciscione^a, Concita Sibilia^a,
Mario Bertolotti^a, Michael Scalora^b, and Mark J. Bloemer^b

^aINFM at Dipartimento di Energetica, Università di Roma "La Sapienza",
Via Scarpa 16, 00161 Roma, Italy;

^bU.S. Army Aviation and Missile Command, Weapon Sciences Directorate, AMSMI-RD-WS-ST
Redstone Arsenal, Huntsville, Alabama 35898-5000

ABSTRACT

We show that inside a multi-layer dielectric stack, consideration of higher-order, fast-oscillating interference terms between counter-propagating waves can dramatically change the dynamics of second harmonic generation, and thus lead to an unusual result: field confinement and overlap can be far better optimized, and conversion efficiencies further enhanced, in the presence of a phase mismatch. One may therefore conclude that phase matching is not always a necessary condition to provide optimized nonlinear frequency conversion efficiency.

Keywords: Non-Linear frequency conversion, multilayer stacks.

Recent work^{1,2} on nanotechnology-based devices has shown that it is possible to design miniaturized structures characterized by a high efficiency/size ratio. The possibility of molding the properties of light using multilayer stacks (or 1D photonic crystals) is of great interest for a wide variety of applications. The technologies covered include waveguides, optical fibers, micro-cavities, channel-drop filters, input/output couplers, all optical multiplexers, and low threshold lasers, to name a few^{3,4}. Additional considerations of nonlinear effects can lead to efficient frequency converters, multicolor sources, and tunable delay lines^{5,6}.

In this paper we focus our attention on materials that display a second order nonlinear response under intense light illumination, more specifically used in devices for second harmonic (SH) generation. As described in reference (6), the fundamental (FF) and SH fields can be made to co-propagate and overlap inside the same layers with roughly the same effective phase index and the same group velocities. Moreover, it has already been shown that in the presence of enhanced local fields resulting from multiple internal reflections, there are terms in the nonlinear polarization that do not conserve momentum^{7,8}. Those terms related to the presence of counter-propagating waves inside the structure are generally neglected in the analysis of nonlinear optical effects because their contribution averages to zero on a length scale of the order of a small fraction of the wavelength.

Although the idea of using multilayer stacks is not new, we wish to show here that it is possible to further increase nonlinear frequency conversion efficiencies by an additional order of magnitude with respect to the state of art. In reference [9] a multiple scale approach was used to derive an analytic expression for the conversion efficiency for a generic layered structure of finite length composed of non-absorbing media. It was found that the conversion efficiency is proportional to the square modulus of an effective coupling coefficient, defined as:

$$\tilde{d}_{eff} = \frac{1}{L} \int_0^L \chi^{(2)}(z) \Phi_{\omega}^2(z) \Phi_{2\omega}^*(z) dz \quad (1)$$

Here L is the length of the structure, and $\Phi_{\omega}(z)$ and $\Phi_{2\omega}(z)$ are the complex, linear field profiles normalized with respect to a unitary input field, so that the electric field inside the structure can be written as: $E_{i\omega}(z) = E_{i\omega}^0 (\Phi_{i\omega}(z) + c.c.)$, where ($i=1,2$), and $E_{i\omega}^0$ is the amplitude of input field. We emphasize that \tilde{d}_{eff} is a complex quantity that contains information

regarding field distribution and localization, as well as contributions to the conversion efficiency coming from the PM conditions. In particular, it is easy to show that for a simple case of SH generation in a bulk medium (i.e. no field localization effects are considered) the square modulus of the coupling coefficient becomes $|\tilde{d}_{eff}|^2 = [\chi^{(2)}]^2 \left[\text{sinc}\left(\frac{\Delta k}{2} L\right) \right]^2$,

where $\Delta k = k(2\omega) - 2k(\omega)$ is the momentum mismatch. Thus, $|\tilde{d}_{eff}|^2$ contains the effect of phase mismatch in the

form of the well-known squared *sinc* function. The concept of maximizing the overlap integral between FF and SH field plays a crucial role in waveguide SH generation¹⁰⁻¹¹. Since the guided-mode is created by interference and localization effects, our arguments find similarities with that approach. However, our model is substantially different because in waveguide configurations the overlap integral is calculated over the transverse coordinate, and then homogenous propagation along the longitudinal axis is assumed. In our case we have longitudinal instead of transverse confinement, which is born out of the multiple reflections that occur as the wave probes the structure. More details can be found in our ref. [9].

As a first step we focus our attention to the fact that our model accurately describes results typically obtained for high-Q doubly-resonant cavities. In particular, the calculation of our \tilde{d}_{eff} in a *homogenous medium* of length L, and refractive index $n(\omega)$, in the presence of counter-propagating waves due to multiple internal reflections can be written as:

$$\tilde{d}_{eff} = \frac{\chi^{(2)}}{L} \left[\int_0^L (A^2 C^* e^{-2ik_o \Delta n z} + B D^* e^{2ik_o \Delta n z}) dz + \int_0^L 2AB (C^* e^{-2ik_o n(2\omega)z} + D^* e^{2ik_o n(2\omega)z}) + (A^2 D^* e^{+4ik_o(n(\omega)+\Delta n/2)z} + B^2 C^* e^{-4ik_o(n(\omega)+\Delta n/2)z}) dz \right] \quad (2)$$

where $\Delta n = n(2\omega) - n(\omega)$ and A,B,C,D are constants that can be calculated by imposing boundary conditions at the input and output interfaces. Indeed, the general expressions for the normalized field profiles inside the medium are: $\Phi_\omega(z) = [A e^{ik_o n(\omega)z} + B e^{-ik_o n(\omega)z}]$ and $\Phi_{2\omega}(z) = [C e^{i2k_o n(2\omega)z} + D e^{-i2k_o n(2\omega)z}]$. In order to get a resonant condition for the FF, L

must be at least of the order of $\lambda/2$. In so choosing L, the fast varying terms inside the integrand average to zero, leaving only the first term on the right hand side (RHS) of Eq (2) to contribute. In spite of local field enhancement, (i.e. magnitudes of A,B,C,D are related to the Q factor of the cavity) the first term of RHS of Eq.(2) shows that the nonlinear dynamics is governed by the PM condition. In other words, due to material dispersion, high conversion efficiency can be achieved only by using birefringent crystals or other PM schemes.

The aim of this letter is to show that the situation dramatically changes when we consider multilayer stacks with layer thicknesses of the order of $\lambda/4$ or less. We consider a structure made by alternating two types of layers, one nonlinear (of high refractive index $n_2(\omega)$) and the other linear ($n_1(\omega)$), of thicknesses δh and δl respectively, so that $\Lambda = \delta h + \delta l$ is the size of the elementary cell, and $L = N\Lambda$, where N is the number of periods. To provide a link between PM conditions and field's overlap, as a first approximation the complex linear field profiles can be decomposed as a superposition of forward and backward waves:

$$\Phi_\omega(z) = [A(z) e^{ik_o n_{eff}(\omega)z} + B(z) e^{-ik_o n_{eff}(\omega)z}]; \text{ and } \Phi_{2\omega}(z) = [C(z) e^{i2k_o n_{eff}(2\omega)z} + D(z) e^{-i2k_o n_{eff}(2\omega)z}]; \quad (3)$$

where n_{eff} is the effective index of refraction as defined in ref. [6], and $A(z)$, $B(z)$, $C(z)$, and $D(z)$, are slowly varying, envelope functions. Substituting Eqs.(3) into the expression for the coupling coefficient, and taking $\chi^{(2)}(z)=0$ everywhere except within the nonlinear layers we obtain:

$$\begin{aligned} \tilde{d}_{eff} = \frac{1}{L} \chi^{(2)} & \left[\sum_{j=0}^N \int_{j\Lambda}^{j\Lambda+\delta h} (A^2(z) C^*(z) e^{-2ik_o \Delta n_{eff} z} + B^2(z) D^*(z) e^{2ik_o \Delta n_{eff} z}) dz + \right. \\ & + 2 \sum_{j=0}^N \int_{j\Lambda}^{j\Lambda+\delta h} A(z) B(z) (C^*(z) e^{-2ik_o n_{eff}(2\omega)z} + D^*(z) e^{2ik_o n_{eff}(2\omega)z}) dz + \\ & \left. + \sum_{j=0}^N \int_{j\Lambda}^{j\Lambda+\delta h} (A^2(z) D^*(z) e^{+4ik_o(n_{eff}(\omega)+\Delta n_{eff}/2)z} + B^2(z) C^*(z) e^{-4ik_o(n_{eff}(\omega)+\Delta n_{eff}/2)z}) dz \right] \quad (4) \end{aligned}$$

where $\Delta n_{eff} = n_{eff}(2\omega) - n_{eff}(\omega)$. The last sum on the right hand side (RHS) of Eq.(4) can be neglected because all terms have a spatial frequency equal to $\pm 2k_{eff}(2\omega)$ ($\cong \pm 4 k_{eff}(\omega)$). We have verified numerically that this term generally gives a contribution much smaller compared to the other terms, unless the integration length (or nonlinear layer thickness) is on the order of $\lambda/8$ or less, where λ is the pump wavelength. However, for such small layer thickness the conversion efficiency has already dropped considerably, and so we choose to optimize conversion rates based on the two leading terms. The first sum on the RHS contains two terms that may be spatially slowly varying depending on the amount of effective phase mismatch. Finally, the terms inside the second sum on the RHS have a spatial frequency equal to $\pm k_{eff}(2\omega)$. Thus, if the optical thickness of the nonlinear layer δh is chosen to be less than a $\lambda/2$ with respect to the SH field (roughly a quarter-wavelength with respect to the pump field) and the spacing (δl) between the nonlinear layers is also properly adjusted, it is then possible to maximize the value of the overall sum in one of two ways, namely, by having either the first or the second term contribute.

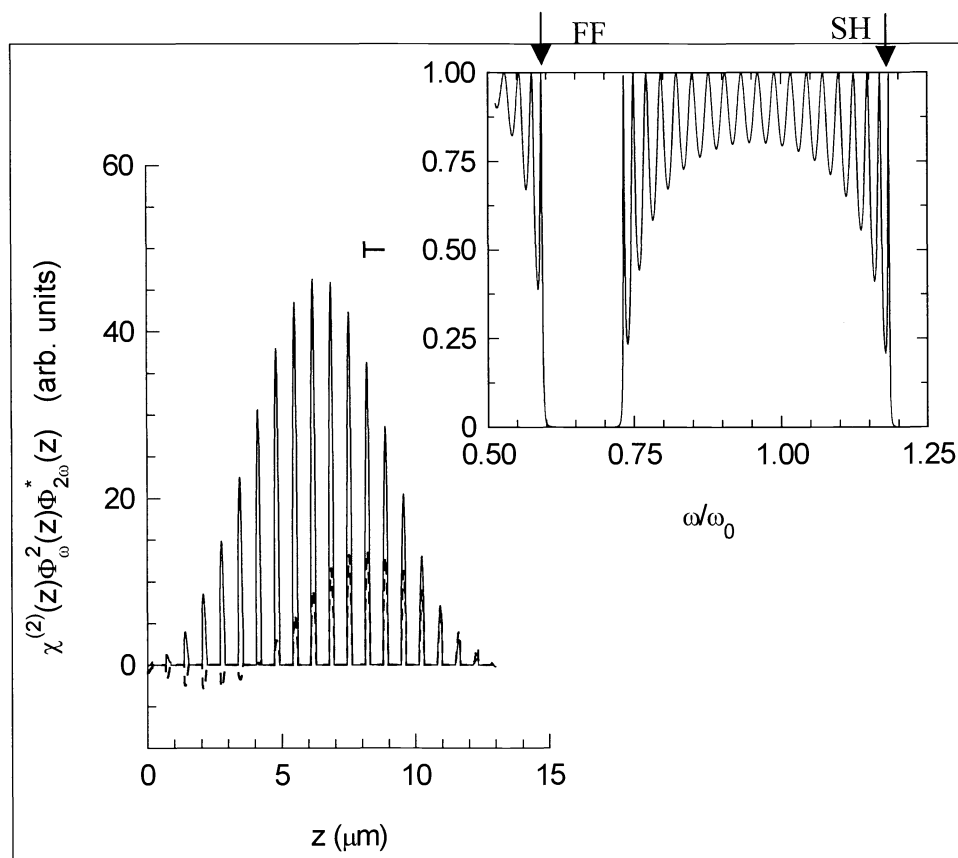


Fig. 1: Real (solid) and Imaginary (dashed) part of the argument of the overlap integral when the FF is tuned at the first band edge resonance, and the SH field is also tuned at the first order band edge resonance.
Inset: transmission spectrum vs frequency at normal incidence. FF and SH tuning are indicated by the arrows.

In Ref.[6] we studied the linear and nonlinear properties of *finite* structures by defining *effective PM conditions* to combine material index dispersion, geometrical dispersion, and field localization. We also showed that the effective PM condition was consistent with the commonly used Bloch wave vector matching for infinite periodic structures. Therefore, we are now claiming that this procedure, as well as all the procedures based on Bloch theory, are equivalent to maximizing the magnitude and importance of *only the first term in Eq.(4)*. Terms due to counter-propagating waves can

indeed enhance frequency conversion rates, in contrast to ordinary rules that apply to standard PM. As an example of a case where conversion efficiency can be further enhanced by at least one order of magnitude compared to a phase matched case we choose a structure with mixed quarter-wave/half-wave geometry and 20 periods. The nonlinear material ($\lambda/4$ optical thickness) has a refractive index $n_2(\omega_{FF})=1.428$ at the FF frequency; for simplicity, the linear material ($\lambda/2$ optical thickness) is assumed to be air, with $n_1=1$. The reference wavelength used to calculate the optical paths of the layers is $1\text{ }\mu\text{m}$, corresponding to an angular frequency $\omega_0=1.88*10^{15}\text{ s}^{-1}$. Assuming normal incidence, this simple geometrical arrangement allows us to rather easily tune the FF and SH fields to the two resonance peaks each located near two consecutive band gaps of the transmission spectrum, where field localization effects are maximized. We tune the FF at the first order band edge resonance ($\lambda_{FF}=1.69\text{ }\mu\text{m}$ which corresponds to $\omega_{FF}=0.592\omega_0$, as labeled by the arrow in the inset of Fig.(1)). Once layer thicknesses have been chosen, one may add dispersion by varying the index of refraction at the SH frequency ($\omega_{SH}=1.184\omega_0$) to tune the field to any desired frequency near the band edge. For example, if dispersion is neglected, $n_2(\omega_{SH})=n_2(\omega_{FF})$, and the SH field is tuned to the 4th resonance transmission peak away from the band edge. Increasing the value of $n_2(\omega_{SH})$ redshifts the band structure, and so it is possible to tune the SH closer to the band edge. Although it is an artifice, varying the refractive index in this fashion makes it possible to find the optimized parameters as a function of one degree of freedom, and to distinguish the role played by phase matching and by local field enhancement during the conversion process. In Fig.(1) we depict the real and imaginary parts of the integrand of Eq. (3) when a suitable index dispersion to the high index material is considered. In particular we have $n_2(\omega_{SH})=1.676$, which effectively tunes the SH to the band edge resonance, as indicated by the arrow in the inset of Fig (1). It is evident that in this case the fast varying contributions to the integral inside the nonlinear layers do not average to zero. Indeed they never change sign inside each nonlinear layer for both real (dashed line) and imaginary (solid) parts. Moreover, all the contributions from each single layer sum coherently to maximize the magnitude of the square modulus of the integral. The expectation of higher conversion efficiencies is fully confirmed by our numerical simulations.

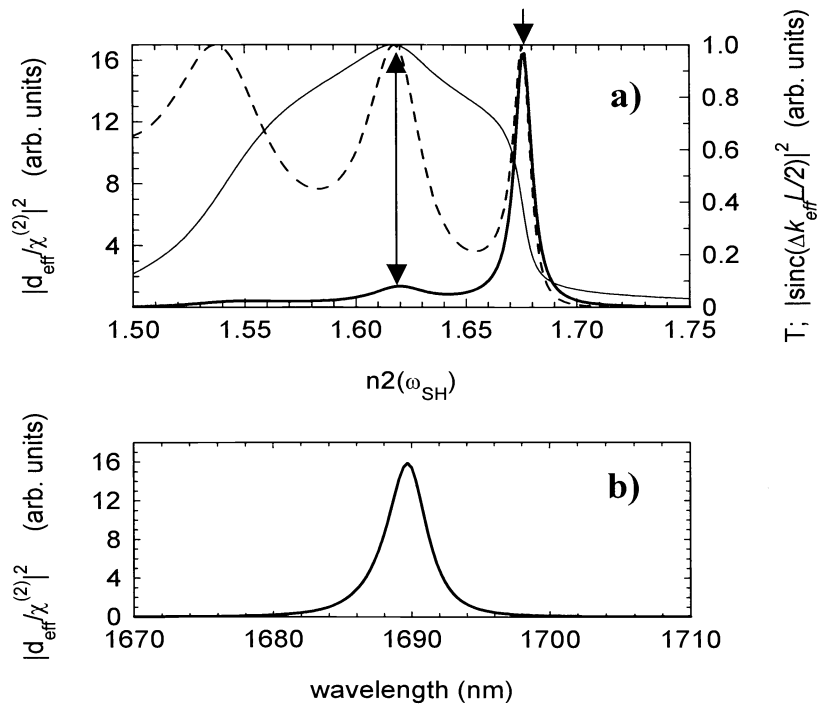


Fig.2: **a)** (thick solid line): Enhancement factor $|\tilde{d}_{eff}/\chi^{(2)}|^2$ vs. $n_2(\omega_{SH})$; (thin solid line): Phase matching contribution given by the expression: $|\text{sinc}(\Delta k_{eff} L/2)|^2$ vs. $n_2(\omega_{SH})$. (right y axis); (dashed line): SH transmission as a function of $n_2(\omega_{SH})$. (left y axis). **b)** $|\tilde{d}_{eff}/\chi^{(2)}|^2$ vs. FF wavelength for the optimized 20 period structure, $\delta h=0.175\text{ }\mu\text{m}$ and $\delta l=0.5\text{ }\mu\text{m}$. The parameters used are $n_1=1$, $n_2(\lambda)=1.333+0.28/\lambda^2-0.025/\lambda^4$ with λ expressed in μm .

In Fig.(2a) we depict the value of the square modulus of $\tilde{d}_{eff} / \chi^{(2)}$ as a function of the refractive index $n_2(\omega_{SH})$ when the pump field remains tuned at the band edge resonance (thick solid line). Changing the refractive index at the frequency ω_{SH} is equivalent to changing the tuning conditions only for the field at ω_{SH} (see transmission spectrum for SH in fig.2a –dashed line). Using only *effective index* considerations, maximum conversion efficiency is expected to be achieved when the second harmonic field is tuned at the second resonance and the ratio $|\tilde{d}_{eff} / \chi^{(2)}|^2$ is described by the function $|\text{sinc}(\Delta k_{eff}L/2)|^2$ [8] plotted in Fig. (2a) (thin line). Comparing the two curves we note that we have only a relative maximum of $|\tilde{d}_{eff} / \chi^{(2)}|^2$ when effective PM is fulfilled. On the other hand, maximum enhancement is achieved when the second harmonic field is tuned to the first band edge resonance, where the effective mismatch is $\Delta n_{eff}=0.06$. We predict that in this case the conversion efficiency will be one order of magnitude greater compared to the case of exact, effective PM. Finally we show the tuning curve ($|\tilde{d}_{eff} / \chi^{(2)}|^2$ vs. FF wavelength) for our optimized structure (Fig 2b). The amount of dispersion introduced, i.e., $n_2(\omega_{SH}) - n_2(\omega_{FF}) = 0.248$, is not atypical of common materials. As a concrete example, an arrangement of 21.5 periods of AlN (140nm)/GaN(94nm) tune the FF at 1068nm, and the SH at 534nm as indicated in the inset of Fig. 1. Here, when $\lambda_{FF}=1.69 \mu\text{m}$, the enhancement factor is more than 3 orders of magnitude larger compared to the out-of-resonance case, with a usable bandwidth of approximately 4nm.

In conclusion, the dynamics of nonlinear frequency conversion is driven by the effective PM conditions only if the field localization is not optimized to enhance the nonlinear interaction. It is possible to achieve higher SH generation conversion efficiency when the fast varying terms in the nonlinear polarization related to the presence of counter-propagating waves are not negligible, and by properly tailoring the size and distribution of the nonlinear layers. The overlap integral plays a crucial role on the conversion efficiency enhancement. It retains the information on effective phase matching but, under appropriate conditions, it can drastically change the dynamics of fields with respect to the case of exact, effective phase matching conditions.

Two of us (M.C. and G.D.) wish to acknowledge the U.S. Army European Research Office for partial financial support.

References:

- 1] E. Yablonovitch, Phys. Rev. Lett. 58, 2059 (1987)
- 2] S. John, Phys. Rev. Lett. 58, 2486 (1987)
- 3] J. Knight, T. Birks, B. Mangan, and P. St. J. Russell, Opt. Photon. News **13**, 26 (2002)
- 4] J. P. Dowling, M. Scalora, M. J. Bloemer, C. M. Bowden, J. Appl. Phys. **75**, 1896 (1994)
- 5] Y. Dumeige, P. Vidakovic, S. Sauvage, I. Sagnes, J. A. Levenson, C. Sibia, M. Centini, G. D'Aguanno, and M. Scalora. Appl. Phys. Lett. **78**, 3021 (2001)
- 6] M. Centini, C. Sibia, M. Scalora, G. D'Aguanno, M. Bertolotti, M. J. Bloemer, C. M. Bowden, and I. Nefedov. Phys. Rev. E **60**, 4891 (1999)
- 7] M. Botey, J. Martorell, J. Trull, R. Vilaseca Opt. Lett. **25**, 1177 (2000)
- 8] A. Yariv, Quantum electronics 3rd edition, John Wiley & Sons; (1989)
- 9] G. D'Aguanno, M. Centini, M. Scalora, C. Sibia, M. Bertolotti, M. J. Bloemer, and C. M. Bowden J. Opt. Soc. Am. B **19**, 2111 (2002)
- 10] J. Khurgin, J. Appl. Phys. **64**, 10 (1988)
- 11] A. Fiore, S. Janz, L. Delobel, P. van der Meer, P. Bravetti, V. Berger, E. Rosencher, J. Nagle Appl. Phys. Lett. **72**, 23 (1998)