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Inverse-designed photonic fibers and metasurfaces for nonlinear frequency conversion [Invited]

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Typically, photonic waveguides designed for nonlinear frequency conversion rely on intuitive and established principles, including index guiding and bandgap engineering, and are based on simple shapes with high degrees of symmetry. We show that recently developed inverse-design techniques can be applied to discover new kinds of microstructured fibers and metasurfaces designed to achieve large nonlinear frequency-conversion efficiencies. As a proof of principle, we demonstrate complex, wavelength-scale chalcogenide glass fibers and gallium phosphide three-dimensional metasurfaces exhibiting some of the largest nonlinear conversion efficiencies predicted thus far, e.g., lowering the power requirement for third-harmonic generation by $10^4$ and enhancing second-harmonic generation conversion efficiency by $10^7$. Such enhancements arise because, in addition to enabling a great degree of tunability in the choice of design wavelengths, these optimization tools ensure both frequency- and phase-matching in addition to large nonlinear overlap factors. © 2018 Chinese Laser Press

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1. INTRODUCTION

Nonlinear frequency conversion plays a crucial role in many photonic applications, including ultra-short pulse shaping [1,2], spectroscopy [3], generation of novel optical states [4–6], and quantum information processing [7–9]. Although frequency conversion has been studied exhaustively in bulky optical systems, including large ring resonators [10] and etalon cavities [11], it remains largely unexplored in micro- and nanoscale structures where light can be confined to length scales of the order of or even smaller than its wavelength. By confining light over long a time and to small volumes, such highly compact devices greatly enhance light–matter interactions, enabling similar as well as new [12] functionalities compared to those available in bulky systems but at much lower power levels. Several proposals have been put forward based on the premise of observing enhanced nonlinear effects in structures capable of supporting multiple resonances at far-away frequencies [13–21], among which are micro-ring resonators [22,23] and photonic crystal (PhC) cavities [24,25]. However, to date, these conventional designs fall short of simultaneously meeting the many design challenges associated with resonant frequency conversion, chief among them being the need to support multiple modes with highly concentrated fields, exactly matched resonant frequencies, and strong mode overlaps [26]. Recently, we proposed to leverage powerful, large-scale optimization techniques (commonly known as inverse design) to allow computer-aided photonic designs that can address all of these challenges.

Our recently demonstrated optimization framework allows automatic discovery of novel cavities that support tightly localized modes at several desired wavelengths and exhibit large nonlinear mode overlaps. As a proof-of-concept, we proposed doubly resonant structures, including multi-layered, aperiodic micro-post cavities and multi-track ring resonators, capable of realizing second-harmonic generation efficiencies exceeding $10^4\text{ W}^{-1}$ [27,28]. In this paper, we extend and apply this optimization approach to design extended structures, including micro-structured optical fibers and PhC three-dimensional metasurfaces, as shown in Fig. 1, for achieving high-efficiency (second- and third-harmonic) frequency conversion. Harmonic generation, which underlies numerous applications in science, including coherent light sources [29], optical imaging and microscopy [30,31], and entangled-photon generation [32], is now feasible at lower power requirements thanks to the availability of highly nonlinear $\chi^{(2)}$ and $\chi^{(3)}$ materials such as III–V semiconductor compounds [33,34] and novel types of chalcogenide glasses [35]. In combination with advances in materials synthesis, emerging fabrication technologies have also
enabled demonstrations of sophisticated micro-structured fibers [36] and metasurfaces [37–44], paving the way for experimental realization of inverse-designed structures of increased geometric and fabrication complexity, which offer orders-of-magnitude enhancements in conversion efficiencies and the potential for augmented functionalities.

Given a material system of intrinsic $\chi^{(2)}$ or $\chi^{(3)}$ nonlinear coefficient, the efficiency of any given frequency-conversion process in a resonant geometry will be determined by a few modal parameters. The possibility of confining light within small mode volumes over a long time or distance leads to significant gains in efficiency (i.e., lower power requirements), stemming from the higher intensity and cascadability of nonlinear interactions (compensating for the otherwise small bulk nonlinearities). In particular, the efficiency of such resonant processes depends on the product of mode lifetimes and a nonlinear coefficient $\beta$, given by Eqs. (6) and (8) below, which generalizes the familiar concept of quasi-phase-matching to situations that include wavelength-scale resonators [26]. For propagating modes, leaky or guided, the existence of a propagation phase further complicates this figure of merit, with optimal designs requiring: (i) phase-matching and frequency-matching conditions, (ii) large nonlinear mode overlaps $\beta$, and (iii) large dimensionless lifetimes $Q$ (low material absorption and/or radiative losses in the case of leaky modes). The main design challenge is the difficult task of forming a doubly resonant cavity with far-apart modes that simultaneously exhibit long lifetimes and large $\beta$, along with phase and frequency matching. To date, the majority of prior works on frequency conversion in fibers [45–47] and metasurfaces [38–40,42,48–51] have focused on only one of these aspects (usually phase matching) while ignoring the others. The geometries discovered by our optimization framework, in contrast, address the above criteria, revealing complex fibers and metasurfaces supporting TE or TM modes with guaranteed phase and frequency matching, long lifetimes $Q$, and enhanced overlap factors $\beta$ at any desired propagation wavevector, and resulting in orders-of-magnitude enhancements in conversion efficiencies.

2. OVERVIEW OF OPTIMIZATION

The possibility of fine-tuning spatial features of photonic devices to realize functionalities not currently achievable by conventional optical design methodologies based on index guiding and bandgap confinement (which work exceedingly well but are otherwise limited for narrowband applications) has been a major drive behind the past several decades of interest in the topic of photonic optimization [52,53]. Among these techniques are probabilistic Monte Carlo algorithms, e.g., particle swarms, simulated annealing, and genetic algorithms [54–56]. Though sufficient for the majority of narrowband (single-mode) applications, many of these gradient-free methods are limited to typically small sets of design parameters [57] that often prove inadequate for handling wideband (multi-mode) problems. On the other hand, gradient-based inverse-design techniques are capable of efficiently exploring a much larger design space by making use of analytical derivative information of the specified objective and constraint functions [58], demonstrated to be feasible for as many as $10^9$ design variables [59]. Recently, the development of versatile mathematical programming methods and the rapid growth in computational power have enabled concurrent progress in photonic inverse design, allowing theoretical (and more recently, experimental) demonstrations of complex topologies and unintuitive geometries with unprecedented functionalities that would be arguably difficult to realize through conventional intuition alone. However, to date, most applications of inverse design in photonics are confined to linear devices such as mode converters, waveguide bends, and beam splitters [57,58,60–65]. We believe that this paper along with our recent works [27,28] provides a glimpse of the potential of photonic optimization in nonlinear optics.

A typical optimization problem seeks to maximize or minimize an objective function $f$, subject to certain constraints $g$, over a set of free variables or degrees of freedom (DOFs) [66]. Generally, one can classify photonic inverse design into two different classes of optimization strategies, based primarily on the nature or choice of DOF [67]. Given a computational domain or grid, the choice of a finite-dimensional parameter space not only determines the degree of complexity but also the convergence and feasibility of the solutions. One possibility is to exploit each DOF in the computational domain as an optimization parameter, known as topology optimization (TO), in which case one typically (though not always) chooses the dielectric permittivity of each pixel $\epsilon(r)$ as a DOF (known as a continuous relaxation parameter [68]). Another possibility, known as shape optimization, is to expand the optimization parameter space in a finite set of shapes (independent of the computational discretization), which may be freeform contours represented by so-called level sets [69] (the level-set method) or basic geometric entities with simpler parametrizations (e.g., polytopes) [70]. In the level-set method, the zeros of a level-set “function” $\Phi(r)$ define the boundaries of “binary shapes”; the optimization then proceeds via a level-set partial differential equation characterized by a velocity field, which is, in turn, constructed from derivative information [69]. A much simpler variant (which we follow) is to choose a fixed but sufficient number of basic binary shapes whose parameters can be made to evolve by an optimization algorithm. Essentially, for such a parametrization, the mathematical representations of the shapes must yield continuous (analytic) derivatives, which is not feasible a priori due to the finite computational discretization and

![Fig. 1. Schematic illustration of third-harmonic generation and second-harmonic generation processes in inverse-designed microstructured fibers and metasurfaces, respectively.](image-url)
can instead be enforced by the use of a “smoothing kernel” (described below).

A generic TO formulation is written down as

\[ \max \int \min f(\epsilon_a), \]

\[ g(\epsilon_a) \leq 0, \]

\[ 0 \leq \epsilon_a \leq 1, \]

where the DOFs are the normalized dielectric permittivities \( \epsilon_a \in [0,1] \) assigned to each pixel or voxel (indexed \( \alpha \)) in a specified volume [58,60]. The subscript \( \alpha \) denotes appropriate spatial discretization \( \mathbf{r} \to (i,j,k)\Delta \) with respect to Cartesian or curvilinear coordinates. Depending on the choice of background (bg) and structural materials, \( \epsilon_a \) is mapped onto a position-dependent dielectric constant via \( \epsilon_a = (\epsilon - \epsilon_{bg}\epsilon_a + \epsilon_{bg}) \). The binarity of the optimized structure is enforced by penalizing the intermediate values \( \epsilon \in (0,1) \) or utilizing a variety of filter and regularization methods [58]. Starting from a random initial guess, the technique discovers complex structures automatically and regularization methods [58]. Starting from a random initial guess, the technique discovers complex structures automatically and regularization methods [58].

For an electromagnetic problem, \( f \) and \( g \) are typically functions of the electric \( \mathbf{E} \) or magnetic \( \mathbf{H} \) fields integrated over some region, which are in turn solutions of Maxwell’s equations under some incident current or field. In what follows, we exploit direct solution of frequency-domain Maxwell’s equations

\[ \nabla \times \frac{1}{\mu} \nabla \times \mathbf{E} - \epsilon(\mathbf{r}, \omega) \mathbf{E} = i\omega \mathbf{J}, \]

describing the steady-state field \( \mathbf{E}(\mathbf{r}, \omega) \) in response to incident currents \( \mathbf{J}(\mathbf{r}, \omega) \) at frequency \( \omega \). While solution of Eq. (4) is straightforward and commonplace, the key to making optimization problems tractable is to obtain a fast-converging and computationally efficient adjoint formulation of the problem. Within the scope of TO, this requires efficient calculations of the derivatives \( \frac{\partial f}{\partial \epsilon_a}, \frac{\partial g}{\partial \epsilon_a} \) at every pixel \( \alpha \), which we perform by exploiting the adjoint-variable method (AVM) [58].

While the TO technique is quite efficient in handling the enormity of an unconstrained design space, it often leads to geometries with irregular features that are difficult to fabricate. An alternative approach that is in principle more conducive to fabrication constraints is to exploit shape optimization. In this work, we primarily focus on a simple implementation of the latter that employs a small and, hence, limited set of elementary geometric shapes, e.g., ellipses [72] and polytopes, parameterized by a few DOFs. In particular, we express the dielectric profile of the computational domain as a sum of basic shape functions with permittivities, \( \epsilon_a = \sum \beta H_\beta(\epsilon_a; \{p_\beta\}) \), described by shape functions \( H_\beta \) and a finite set of geometric parameters \( \{p_\beta\} \), where \( \beta \) denotes the shape index. Here, to deal with potential overlap of two or more shapes, we implement a filter function that enforces the same maximum-permittivity constraint \( \epsilon \leq 1 \) described above. The derivatives of a given objective function \( f \) (and associated constraints) can then be obtained via the chain rule \( \frac{\partial f}{\partial p_\beta} = \frac{\partial f}{\partial \epsilon_a} \frac{\partial \epsilon_a}{\partial p_\beta} \), where the smoothness of the derivatives is guaranteed by insisting that the shape functions \( H \) be continuously differentiable functions. Below, we choose non-piecewise-constant ellipsoidal shapes with exponentially varying dielectric profiles near the boundaries, the smoothness of which is determined by a few simple parameters that can, at various points along the optimization, be slowly adjusted to realize fully binary structures upon convergence. Such a “relaxation” process [70] is analogous to the application of a binary filter in the objective function [58].

Any nonlinear frequency conversion process can be viewed as a frequency mixing scheme in which two or more constituent photons at a set of frequencies \( \{\omega_n\} \) interact to produce an output photon at frequency \( \Omega = \sum \omega_n \omega_m \) where \( \{\epsilon_n\} \) can be either negative or positive, depending on whether the corresponding photons are created or destroyed in the process [73]. Given an appropriate nonlinear tensor component \( \chi_{ijkl} \), with \( i,j,k,\ldots \in \{x,y,z\} \), mediating an interaction between the field components \( E_i(\Omega) \) and \( E_{jk}, E_{k\ell}, \ldots \), we begin with a collection of point dipole currents, each at the constituent frequency \( \omega_n, n \in \{1,2,\ldots\} \), such that \( J_n = \hat{\epsilon}_n \delta(\mathbf{r} - \mathbf{r}') \), where \( \hat{\epsilon}_n \in \{\hat{e}_1, \hat{e}_2, \ldots\} \) is a polarization vector chosen so as to excite the desired electric-field polarization components \( \hat{\epsilon}(\Omega) \) of the corresponding mode at an appropriate position \( \mathbf{r}' \).

Given the choice of incident currents \( J_n \), we solve Maxwell’s equations to obtain the corresponding constituent electric-field response \( \mathbf{E}_n \), from which one can construct a nonlinear polarization current \( J(\Omega) = \hat{\epsilon}_n(\mathbf{r}) |_{\mathbf{E}_n} \), where \( \mathbf{E}_n = \hat{\epsilon}_n \cdot \hat{\epsilon}_n \) and \( J(\Omega) \) can be generally polarized \( \hat{\epsilon}(\Omega) \) in a (chosen) direction that differs from the constituent polarization \( \hat{\epsilon}_n \). Here, \( (\ast) \) denotes complex conjugation for negative \( \epsilon_n \) and no conjugation otherwise. Finally, maximizing the radiated power, \( \text{Re} \left[ \int J(\Omega)^* \cdot \mathbf{E}(\Omega) \mathbf{d}r \right] \), due to \( J(\Omega) \), one is immediately led to the following nonlinear optimization problem:

\[ \max_{\mathbf{E}} f(\epsilon; \omega_n) = -\text{Re} \left[ \int J(\Omega)^* \cdot \mathbf{E}(\Omega) \mathbf{d}r \right], \]

\[ \mathcal{M}(\epsilon, \omega_n) \mathbf{E}_n = i\omega \mathbf{J}_n = \hat{\epsilon}_n \delta(\mathbf{r} - \mathbf{r}') , \]

\[ \mathcal{M}(\epsilon, \Omega) \mathbf{E}(\Omega) = i\Omega J(\Omega), \]

\[ J(\Omega) = \hat{\epsilon}_n(\mathbf{r}) |_{\mathbf{E}_n} \]

\[ \mathcal{M}(\epsilon, \omega, \Omega) = \nabla \times \frac{1}{\mu} \nabla \times -\epsilon(\mathbf{r}, \omega) \mathbf{E}, \]

where \( \epsilon \) is given by either the topology or shape parameterizations described above. Writing down the objective function in terms of the nonlinear polarization currents, it follows that solution of Eq. (5), obtained by employing any mathematical programming technique that makes use of gradient information, e.g., the AVM [58], maximizes the nonlinear coefficient (mode overlap) associated with the aforementioned nonlinear optical process. The above framework can be easily extended to consider propagating modes once we take into account the appropriate Bloch boundary conditions that may arise from any desired wave vectors imposed at the requisite frequencies [74]. In the case of optical fibers or PhC metasurfaces (or, more generally, any waveguiding system), such an extension naturally guarantees perfect phase and frequency matching of the relevant modes in the optimized structure.

3. THIRD-HARMONIC GENERATION IN FIBERS

Conventional microstructured fibers (e.g., Bragg and holey fibers) are typically designed based on intuitive principles like
slow light [47], index guiding, and bandgap confinement [52], and thus often consist of periodic cross sections comprising simple shapes [75,76]. Below, we apply the aforementioned optimization techniques to propose much more complicated heterostructure fibers designed to enhance third-harmonic generation at any desired wavelength. To achieve large third-harmonic generation efficiencies, the fiber must support two co-propagating modes of frequencies $\omega_1$ and $\omega_3 = 3\omega_1$ and wavenumbers that satisfy the phase-matching condition $k_3 = 3k_1$. Furthermore, the system must exhibit low radiative/dissipative losses or, alternatively, attenuation lengths that are much longer than the corresponding interaction lengths $L$, defined as the propagation length at which 50% of the fundamental mode is upconverted. In the small-input signal regime, the converted third-harmonic output power $P_3 \propto P_1^2$ and the interaction length $L = \frac{16}{3\kappa Z_0 P_3}$ depend on the incident power $P_1$, vacuum impedance $Z_0$, and nonlinear overlap factor $\beta_3$.

\[
\beta_3 = \frac{24\pi}{\sqrt{\text{Re}\left[\frac{1}{2} \sqrt{\left(\mathbf{E}_1 \cdot \mathbf{H}_1\right)} \cdot 2dS\right]^3}} \left|\text{Re}\left[\frac{1}{2} \sqrt{\left(\mathbf{E}_3 \cdot \mathbf{H}_3\right)} \cdot 2dS\right]\right|
\]

(6)

which involves a complicated spatial overlap of the two modes over the cross-sectional surface $S$ of the fiber. Note that the attenuation coefficient $\gamma \equiv \omega_1/2\pi Q$ of each mode (the inverse of their respective attenuation length) is proportional to their lifetime $Q$ and group velocity $v_g$.

We focus on fibers comprising chalcogenide/polyethersulfone (PES) composites of permittivities $\epsilon_{\text{ch/S}} = 5.8125$ and $\epsilon_{\text{PES}} = 2.4025$ at telecom wavelengths. Although our technique can be readily applied to design the requisite properties at any given wavenumber $k$ and for any desired polarization, we specifically focus on designs for operation at wavenumbers in the range $0.1(2\pi/\lambda) < k_{\text{opt}} < 3.2(2\pi/\lambda)$, with $\lambda$ denoting the corresponding vacuum wavelength and $k_{\text{opt}}$ the optimized wavenumber. We consider both leaky and guided modes above and below the PES lightline $\omega = c k/\sqrt{\epsilon_{\text{PES}}}$, respectively, along with different choices or transverse electric TE$_{01}$ and transverse magnetic TM$_{01}$ polarizations. TE$_{01}$ modes are those polarized along the plane of the fiber and consist primarily of circulating $E_x$ and $E_y$ electric fields [78], while TM$_{01}$ modes have electric fields $E_z$ polarized mainly along the propagation direction $z$.

The top insets in Fig. 2 show an inverse-designed fiber cross section that supports phased-matched TM$_{01}$ fundamental and third-harmonic modes (with profiles superimposed on the inset) at $k_{\text{opt}} = k_1 = 1.4(2\pi/\lambda)$. To ensure that the optimization algorithm selectively finds TM$_{01}$ modes, we employ a magnetic current $\mathbf{J}_1 \sim \nabla \times \mathbf{\delta}(r) \mathbf{e}_z$ as the source in Eq. (5), resulting in electric fields of the desired polarization. The fiber cross section is represented by a $34 \times 34$ computational cell consisting of 300 pixels × 300 pixel, where the size of each pixel is 0.01$\lambda$ × 0.01$\lambda$. From Fig. 2 (inset), it is clear that both the fundamental and third-harmonic modes are well confined to the fiber core and exhibit substantial modal overlaps, while again, the phase-matching condition is automatically satisfied by the optimization process, with $k_3 = 3k_1$. We find that $|\beta_3|^2 \approx 2 \times 10^4 (\chi^{(3)}/\lambda)$ is almost 4 orders of magnitude larger than what has been demonstrated in standard plain fibers, which have typical values of $|\beta_3|^2 \lesssim 2 (\chi^{(3)}/\lambda^2)$ [77]. Figure 2 shows the dispersion of the two leaky modes (solid lines), with the PES lightline represented by the gray region and their corresponding dimensionless lifetimes, around $Q_1 \approx 10^2$ and $Q_3 \approx 10^3$ at $k_{\text{opt}}$, plotted as dashed lines. Noticeably, while the fiber is optimized to ensure phase matching at a single $k_{\text{opt}}$, any phase mismatch remains small in the vicinity of $k \approx k_{\text{opt}}$. In fact, even for $k \ll k_{\text{opt}}$, the frequency difference is found to be only around 1%. Technically, the only factor limiting the lifetimes is the finite computational cross-section (imposed by the finite computational cell), with much larger lifetimes possible for larger cross sections. Away from $k_{\text{opt}}$, the quality factors decrease while remaining relatively large over a wide range of $k$.

Considering the group velocity $v_g$ around $k_{\text{opt}}$, we find that the attenuation length of the fiber $L_{\text{rad}} = 1/\gamma \approx 2v_g Q/\omega \approx 1.66 \times 10^3 \lambda$. We note that while the fiber supports multiple modes around these wavelengths, the only modes near $k_{\text{opt}}$ are those discovered by the optimization and shown in the figure.

Figure 3 shows the $\beta_3$ corresponding to fibers optimized for operation at different values of $k_{\text{opt}}$ and polarizations, and obtained by application of either topology (squares or circles) or shape (triangles) optimization. The figure shows a general trend in which $\beta_3$ decreases with increasing $k_{\text{opt}}$ for both polarizations, except that TM$_{01}$ fibers tend to exhibit non-monotonic behavior, with $\beta_3$ increasing sharply at an intermediate $k_{\text{opt}} \approx 2\pi/\lambda$ below the lightline, above which it drops significantly before increasing again in the guided regime, peaking again at $k_{\text{opt}} \approx 1.7(2\pi/\lambda)$ before plummeting once again. We suspect that this complicated behavior is not a consequence of any
METASURFACES

4. SECOND-HARMONIC GENERATION IN METASURFACES

Metasurfaces offer an advantageous platform for realizing complicated beam generation and wavefront shaping over extended surfaces [79] and have recently been exploited in conjunction with nonlinear materials as a means of generating and controlling light at multiple wavelengths [43,48,80,81]. A typical nonlinear metasurface can suffer from poor frequency-conversion efficiencies due to a combination of weak confinement, material absorption, and sub-optimal mode overlaps. In particular, typical designs exploit plasmonic [38–40,50] or all-dielectric [42,49] elements comprising simple shapes distributed over a unit cell, including split ring resonators [38,40,50], cross bars [39], and cylindrical posts [49], with the main focus being that of satisfying the requisite frequency- and phase-matching condition [82]. Here, we show that inverse design can not only facilitate the enforcement of frequency- and phase-matching requirements but also allow further enhancements stemming from the intentional engineering of nonlinear modal overlaps, often neglected in typical designs.

To achieve large second-harmonic generation efficiencies, a metasurface must support two extended resonances at frequencies \( \omega_1 \) and \( \omega_2 = 2 \omega_1 \) and wavevectors satisfying the phase-matching condition \( k_3 = 2k_1 \). As illustrated schematically in Fig. 4(a), a typical setup consists of an incident wave of power per unit cell \( P_1 \) at some frequency and angle (described by wavenumber \( k_1 \)) and a corresponding output harmonic wave of power per unit cell, \( P_2 \). In the small-signal regime, the output power \( P_2 \propto P_1^2 \) scales quadratically with \( P_1 \), resulting in a conversion efficiency per unit cell of

\[
\eta = \frac{P_2}{P_1} = \frac{Q_1^2 Q_2^2}{Q_{1,rad} Q_{2,rad}} \frac{|\beta_3|^2 \lambda_1}{\pi \epsilon_0 \epsilon},
\]

where \( Q \) and \( Q_{\text{rad}} \) denote total and radiative dimensionless lifetimes and \( \beta_3 \) the nonlinear overlap factor:

\[
\beta_3 = \frac{\int d\mathbf{V} \chi^{(3)} \mathbf{E}^*_i \cdot \mathbf{E}_i}{\left( \int d\mathbf{V} |\mathbf{E}_i|^2 \right)^2}. 
\]

Note that here the conversion efficiency is defined as the efficiency per unit cell for such an extended surface, hence the volume integration is performed inside a unit cell.

We now apply our optimization framework to discover new all-dielectric three-dimensional metasurfaces, with the permittivity of the medium \( \epsilon_{\text{gap}} \) taken to be that of gallium phosphide near telecom wavelengths [83,84]. Note, however, that the same framework can be easily extended to design plasmonic...
The optimized designs exhibit orders of magnitude larger modal parameters in these studies [38, 39, 49, 50], such as the

**radiative lifetimes** $Q_{\text{rad}} = 6(2) \times 10^4$ and overlap factor $|\beta_2|^2 = 1.6 \times 10^{-3}(\chi^{(2)}/\lambda^3)$. The structure on the right is...


