Phase-matching solutions for high-order harmonic generation in hollow-core photonic-crystal fibers

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Hollow-core photonic-crystal fibers are shown to allow phase-matched high-order harmonic generation by an isolated guided mode of pump radiation. Regimes of phase matching are analyzed for the fundamental guided mode of pump field with a wavelength around 800 nm, generating harmonics within the wavelength range of 25–50 nm in hollow photonic-crystal fibers filled with argon, krypton, and helium. Geometric parameters of the fiber structure and the pressure of the gas filling the fiber core are shown to serve as important, often orthogonal, control knobs, allowing a fine adjustment of the phase matching for high-order harmonic generation.

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I. INTRODUCTION

Hollow-core fibers [1,2] offer interesting new options for high-field ultrafast nonlinear optics. Such fibers can guide laser pulses with intensities up to the plasma-formation threshold with no damage to the waveguide itself [3] and can serve to increase the effective interaction length, allowing a radical enhancement of nonlinear-optical processes, including four-wave mixing [4–6] and high-order harmonic generation [7–10]. Hollow fibers have been shown to allow efficient compression and chirp control of high-energy ultrashort laser pulses due to the Kerr-nonlinearity-induced self-phase modulation (SPM) [11,12] and high-order stimulated Raman scattering (SRS) [13].

Air-guided modes in standard hollow fibers are, however, leaky, with the magnitude of losses scaling as [1,2] λ^2/a^3 with the fiber inner radius a and the radiation wavelength λ, which dictates the choice of hollow fibers with $a \sim 50-300\ \mu$m for nonlinear-optical experiments. Such large-a fibers are essentially multimode, with many guided modes typically contributing to nonlinear-optical interactions [6], sometimes leading to unwanted interference phenomena in wave-mixing processes [14].

Hollow-core photonic crystal fibers (PCFs) [15,16] resolve this conflict between the magnitude of radiation losses and the number of air-guided modes. PCFs guide light due to the high reflectivity of a two dimensionally periodic (photonic-crystal) cladding (the inset in Fig. 1) within photonic band gaps (PBGs). Low-loss guiding in a few or even a single air-guided mode can be implemented under these conditions in a hollow core with a typical diameter of 10–20 μm [15–20]. Hollow PCFs with such core diameters have been recently demonstrated to enhance nonlinear-optical processes, including stimulated Raman scattering [21], four-wave mixing (FWM) [22], and self-phase modulation [23]. The spatial self-action of intense ultrashort laser pulses gives rise to interesting waveguiding regimes in hollow PCFs below the blowup threshold [24]. The SPM-induced spectral broadening of femtosecond laser pulses in air-guided modes of hollow PCFs allows the creation of fiber-optic diodes [23] and limiters [25] for high-intensity ultrashort laser pulses. Air-guided modes in hollow PCFs can support megawatt optical solitons [26] and allow femtosecond soliton pulse delivery over several meters [27], as well as transportation of high-energy laser pulses for technological [28,29] and biomedical [30] applications.

Demonstration of the whole catalog of $\chi^{(3)}$-controlled nonlinear-optical processes (viz., SRS [21], FWM [22], SPM [23], soliton formation [26,27], and spatial self-action [24]) radically enhanced in hollow PCFs raises an interesting question as to whether enhancement strategies offered by hollow PCFs can be extended to higher-order nonlinear processes, which have been recently shown to allow the genera-

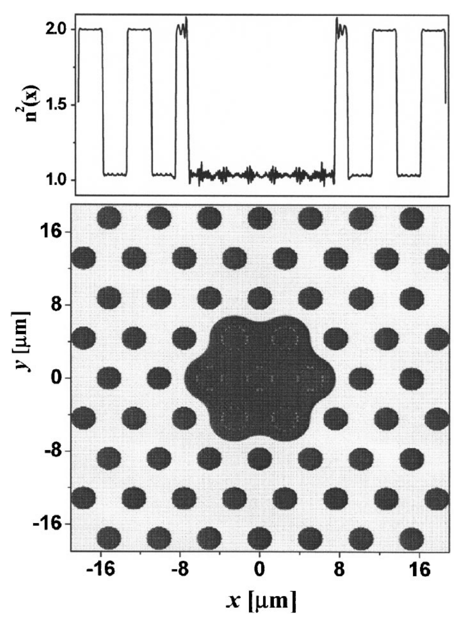

FIG. 1. The profile of $n^2(x,y)$ in the cross section of a hollow PCF synthesized with 80×80 Hermite-Gaussian polynomials and 150×150 trigonometric functions: (top) 1D cut, (bottom) 2D profile shown by levels of gray scale.

tion of a few femtosecond and attosecond pulses [31–33]. Standard hollow fibers have been already used for the enhancement of high-order harmonic generation [7–10] and multiple stimulated Raman scattering [13]. Dispersion of air-guided modes in hollow PCFs, however, can generally quite substantially differ from the mode dispersion of standard hollow fibers. In this work, we address this issue by analyzing phase-matching solutions for high-order harmonic generation in hollow PCFs. We will show that hollow PCFs allow phase-matched high-order harmonic generation by an isolated air-guided mode of pump radiation. We will illustrate this possibility by studying the generation of high-order harmonics within the wavelength range of 25–50 nm by the fundamental air-guided mode of 800-nm pump field in a rare-gas-filled hollow PCF.

II. WAVEGUIDE MODES OF A HOLLOW PHOTONIC-CRYSTAL FIBER

To model waveguide modes of a hollow PCF, we developed a numerical procedure solving the vectorial wave problem for the transverse components of the electric field $\vec{E}(z,t)=\vec{E}\exp[i(\beta z-ckt)]$, where $\vec{E}=(E_x,E_y,E_z)$, β is the propagation constant, $k=\omega/c$ is the wave number, ω is the radiation frequency, and c is the speed of light,

$$\left[\frac{\nabla_\perp^2}{k^2}+n^2(x,y)\right]\mathrm{E}_x+\frac{1}{k^2}\frac{\partial}{\partial x}\left(\mathrm{E}_x\frac{\partial\ln(n^2)}{\partial x}+\mathrm{E}_y\frac{\partial\ln(n^2)}{\partial y}\right)=\frac{\beta^2}{k^2}\mathrm{E}_x, \quad (1)$$

$$\left[\frac{\nabla_\perp^2}{k^2}+n^2(x,y)\right]\mathrm{E}_y+\frac{1}{k^2}\frac{\partial}{\partial y}\left(\mathrm{E}_x\frac{\partial\ln(n^2)}{\partial x}+\mathrm{E}_y\frac{\partial\ln(n^2)}{\partial y}\right)=\frac{\beta^2}{k^2}\mathrm{E}_y. \quad (2)$$

Here, $\nabla_\perp$ is the gradient in the (x,y) plane, $n=n(x,y)$ is the two-dimensional profile of the refractive index, and the prime in the second term on the left-hand side stands for differentiation. The profile of $n^2(x,y)$ was approximated, similar to [34,35], with a series expansion in Hermite-Gaussian polynomials and trigonometric functions. Figure 1 displays a 1D cut (top) and a 2D profile (bottom) of $n^2(x,y)$ in the PCF cross section, synthesized with 80×80 Hermite-Gaussian polynomials and 150×150 trigonometric functions. The E_x and E_y components of the electric field were represented as series expansions in Hermite-Gaussian polynomials. Analysis of phase matching for high-order harmonic generation will be performed in this work for PCFs with a period of the cladding Λ equal to 5 and 10 μm. A substitution of the series expansions for E_x, E_y, and $n^2(x,y)$ into Eqs. (1) and (2) reduces the problem to an eigenfunction and eigenvalue problem of a matrix equation, which allows the propagation constants and transverse field profiles to be determined for the air-guided modes of hollow PCFs.

Interestingly and very importantly for phase-matched high-order harmonic generation, different physical mechanisms are responsible for the guiding of the pump and harmonic fields in the gas-filled hollow core of a PCF. With the refractive index of the gas filling the PCF core being lower than the refractive index of the material of the PCF cladding at the frequency of pump radiation, the pump field can be guided only by PBGs in PCFs with a small core diameter, leaking out of the hollow core if the frequency of the pump field is tuned off the PBGs of the cladding. The relation between the refractive indices of the gas filling the PCF core and the material of the PCF cladding becomes opposite at frequencies of high-order harmonics, which can be therefore guided by total internal reflection at any gas pressure.

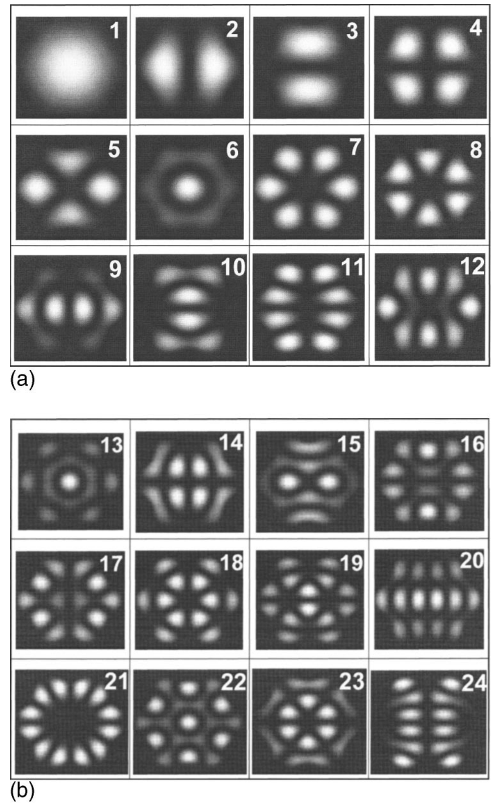

FIG. 2. Intensity profiles in the guided modes of a hollow PCF. The mode index j, shown in the upper right corner, ranges from 1 to 12 (a) and from 13 to 24 (b).

Figure 2 presents typical field intensity profiles for the fundamental and higher-order guided modes in a hollow PCF calculated with the use of the numerical procedure solving Eqs. (1) and (2) as described above. The fundamental mode (mode 1 in Fig. 2) has the maximum effective refractive index $n_{eff}=\beta/k$, and the electric-field intensity for this mode reaches its maximum at the center of the fiber core, monotonically decreasing off the center of the fiber. Higher-order modes form degenerate multiplets, with their superposition supporting the full symmetry of the fiber [36]. In what follows, we will explore the possibilities of phase-matched generation of the fundamental and higher-order modes of optical harmonics by the fundamental mode of the pump field.

We consider phase matching for the generation of high-order harmonics with wavelengths falling within the range of 25–50 nm by pump radiation with a wavelength around

800 nm. The number of modes of the pump field guided by the hollow core of the PCF is controlled [15,37] by the maximum core wave vector, $k_1=\omega n_1/c$, where n_1 is the refractive index of the gas in the fiber core. This implies that the gas pressure in the PCF core is an important factor controlling the number of air-guided modes of the pump field in a hollow PCF. Although a hollow PCF with the structure of cross section shown in Fig. 1 and the period of the cladding Λ ranging from 5 to 10 μm is not truly a single mode for 800-nm pump radiation, an isolated fundamental guided mode can be easily excited within the wavelength range around 800 nm with a properly coupled input field [38], allowing high-order harmonic generation by a pump field with a well-defined transverse intensity profile.

III. PHASE-MATCHED HIGH-ORDER HARMONIC GENERATION IN HOLLOW PHOTONIC-CRYSTAL FIBERS: GENERAL STRATEGY

High efficiencies of nonlinear-optical interactions can be achieved only when the phase of the field generated as a result of the nonlinear optical process is matched with the phase of the nonlinear polarization induced in the medium by the pump field. In the case of qth-harmonic generation in an optical waveguide, this condition, generally known as the phase-matching condition in nonlinear optics [39], can be written in terms of the propagation constants β_m and β_n of the mth guided modes of the harmonic and the nth mode of the pump field,

$$\beta_m(q\omega_p)=q\beta_n(\omega_p), \tag{3}$$

where ω_p is the frequency of the pump field.

The phase-matching condition (3) requires the equality of the effective refractive indices $n_{eff}=\beta c/\omega$ (where c is the speed of light) at the wavelengths of the pump field and its qth harmonic. In Figs. 3–6, we explore the ways of matching the effective refractive index of the fundamental mode of pump radiation with a wavelength around 800 nm and the effective refractive index of the fundamental (Figs. 3–5) and higher-order (Fig. 6) modes of high-order harmonics emitted within the range of wavelengths from 25 up to 50 nm for hollow PCFs filled with argon and krypton. The upper horizontal scale in these figures represents the pump wavelength λ_p, while the lower horizontal scale displays the wavelength λ_h of harmonic emission. Phase matching is achieved where the dispersion curve of the fundamental waveguide mode of the pump field crosses the dispersion curve of one of the guided modes of high-order harmonics.

IV. PHASE-MATCHED HIGH-ORDER HARMONIC GENERATION IN HOLLOW PHOTONIC-CRYSTAL FIBERS: THE EFFECT OF THE GAS PRESSURE AND FIBER STRUCTURE

Figures 3–5 illustrate the influence of the gas pressure in the fiber core and the structure of the PCF on phase matching in high-order harmonic generation. While Figs. 3 and 4 demonstrate the possibility to minimize the phase mismatch for a whole group of high-order harmonics of the pump field, Fig.

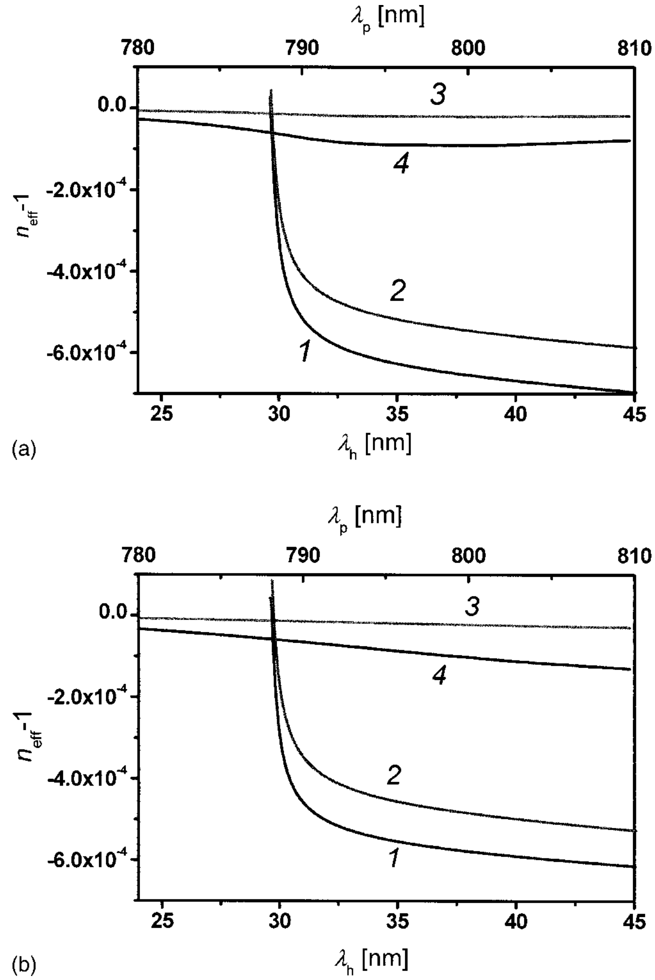

FIG. 3. Effective refractive indices for the fundamental modes of the pump field (1, 2) and high-order harmonics (3, 4) shown as functions of the pump and harmonic wavelengths λ_p and λ_h (the upper and lower horizontal axes, respectively) for a hollow PCF with $\Lambda=5$ μm filled with (a) argon and (b) krypton at a pressure of (1, 3) 0.1 and (2, 4) 0.5 atm.

5 presents the difference of effective refractive indices $\delta n_q=n_{\text{eff}}(q\omega)-n_{\text{eff}}(\omega)$ for the fundamental mode of the pump field and the fundamental mode of a specific harmonic [$q=21$ in Figs. 5(a) and 5(b), and $q=19$ in Fig. 5(c)], showing the conditions for perfectly phase-matched qth harmonic generation by a pump field with a wavelength of approximately 788 nm in argon- and krypton-filled hollow PCFs with different Λ. The gas pressure is varied within the range from 0.05 atm up to 0.5 atm in our calculations, with a typical absorption length for 25-nm radiation being on the order of 1 cm for 0.1 atm of argon or krypton.

The phase mismatch and the wavelength of exact phase matching can be finely adjusted, as can be seen from the results of our calculations, by varying the gas pressure and changing the period of the PCF cladding Λ. The data presented in Figs. 3–5 show that these two adjustments are nearly orthogonal for the considered regime of harmonic generation. Since the material dispersion plays a much more important role for short-wavelength radiation of high-order harmonics, gas-pressure variations, as can be seen from Figs.

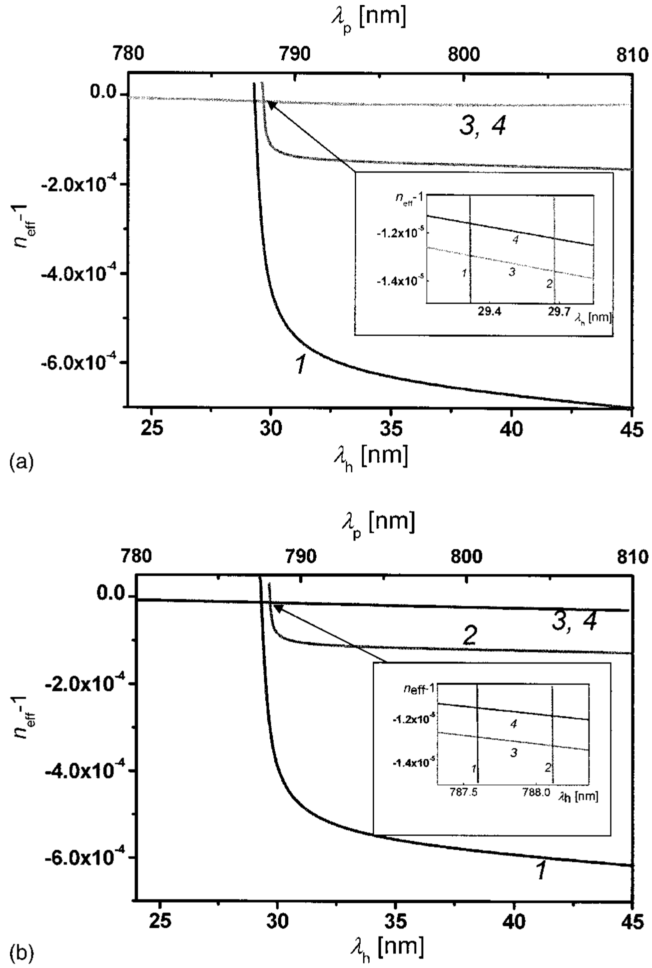

FIG. 4. Effective refractive indices for the fundamental modes of the pump field (1, 2) and high-order harmonics (3, 4) shown as functions of the pump and harmonic wavelengths (the upper and lower horizontal axes, respectively) for a hollow PCF with $\Lambda = 5\ \mu$m (1, 3) and 10 μm (2, 4) filled with (a) argon and (b) krypton at a pressure of 0.1 atm. The insets show the details of the phase-matching regions.

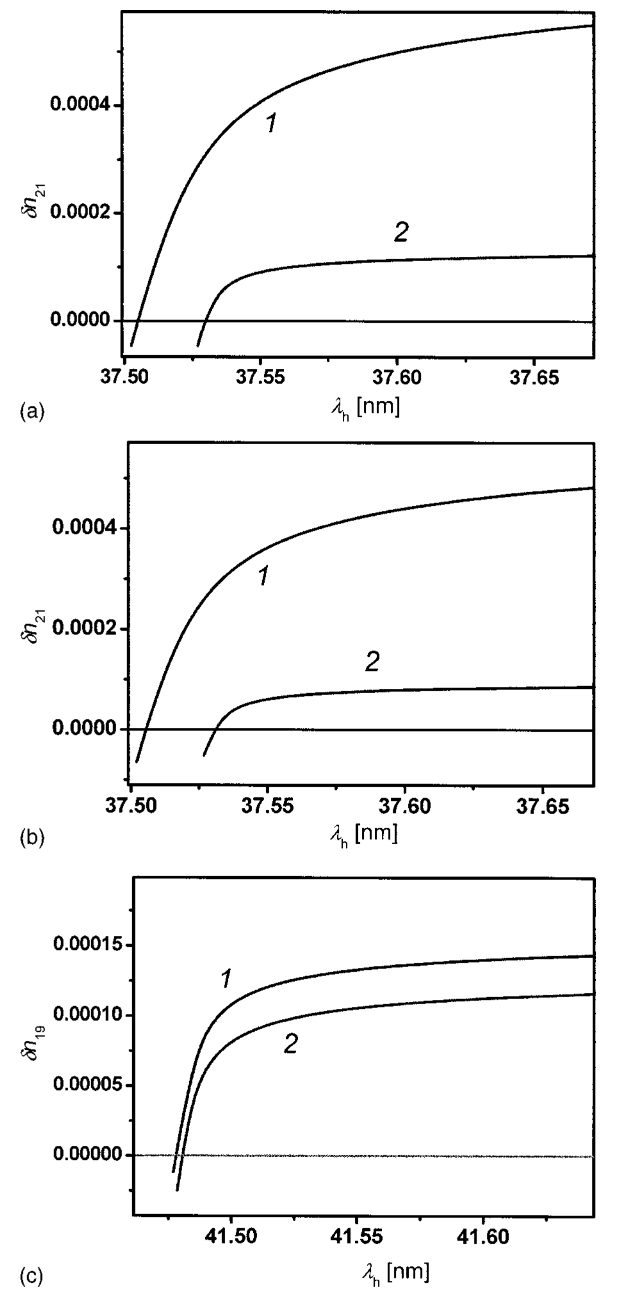

FIG. 5. The difference of effective refractive indices $\delta n_q = n_{\text{eff}}(q\omega) - n_{\text{eff}}(\omega)$ for the fundamental mode of the pump field and the fundamental mode of the qth harmonic as a function of the harmonic wavelength in an argon- or krypton-filled hollow PCF: (a) $q=21$, a hollow PCF filled with argon at a pressure of 0.1 atm with (1) $\Lambda=5$ and (2) $\Lambda=10\ \mu$m; (b) $q=21$, a hollow PCF filled with krypton at a pressure of 0.1 atm with (1) $\Lambda=5$ and (2) $\Lambda=10\ \mu$m; and (a) $q=19$, a hollow PCF filled with argon (1) and krypton (2) at a pressure of 0.05 atm with $\Lambda=10\ \mu$m.

3(a) and 3(b), can noticeably change the effective refractive index of the waveguide mode of the harmonic field, having virtually no impact on the behavior of the bending section of the dispersion of the pump field. The guided modes of harmonic emission, on the other hand, are not very sensitive to changes in the PCF structure, since harmonic wavelengths are much less than the fiber core. Dispersion of the guided mode of the pump field, at the same time, can be efficiently tuned by changing the fiber structure [Figs. 4(a), 4(b), 5(a), and 5(b)]. Gas-pressure and fiber-structure variations can thus serve as orthogonal control knobs for independent fine adjustments of the pump-field dispersion versus the dispersion of the harmonic field, as well as for switching the number of the optical harmonic corresponding to the minimum phase mismatch.

The wavelength of optical harmonics remains much less than the fiber core diameter even for hollow PCFs with a core diameter as small as 10–20 μm. Phase-matching the fundamental guided modes of the pump field and the field of one of the harmonics should therefore automatically result in phase-matched harmonic emission in several waveguide modes. This aspect of harmonic generation in hollow PCFs is illustrated in Figs. 6(a)–6(e). Changes in the fiber structure, the sort of gas, and the gas pressure, as can be seen from these plots, can help to optimize phase-matching conditions

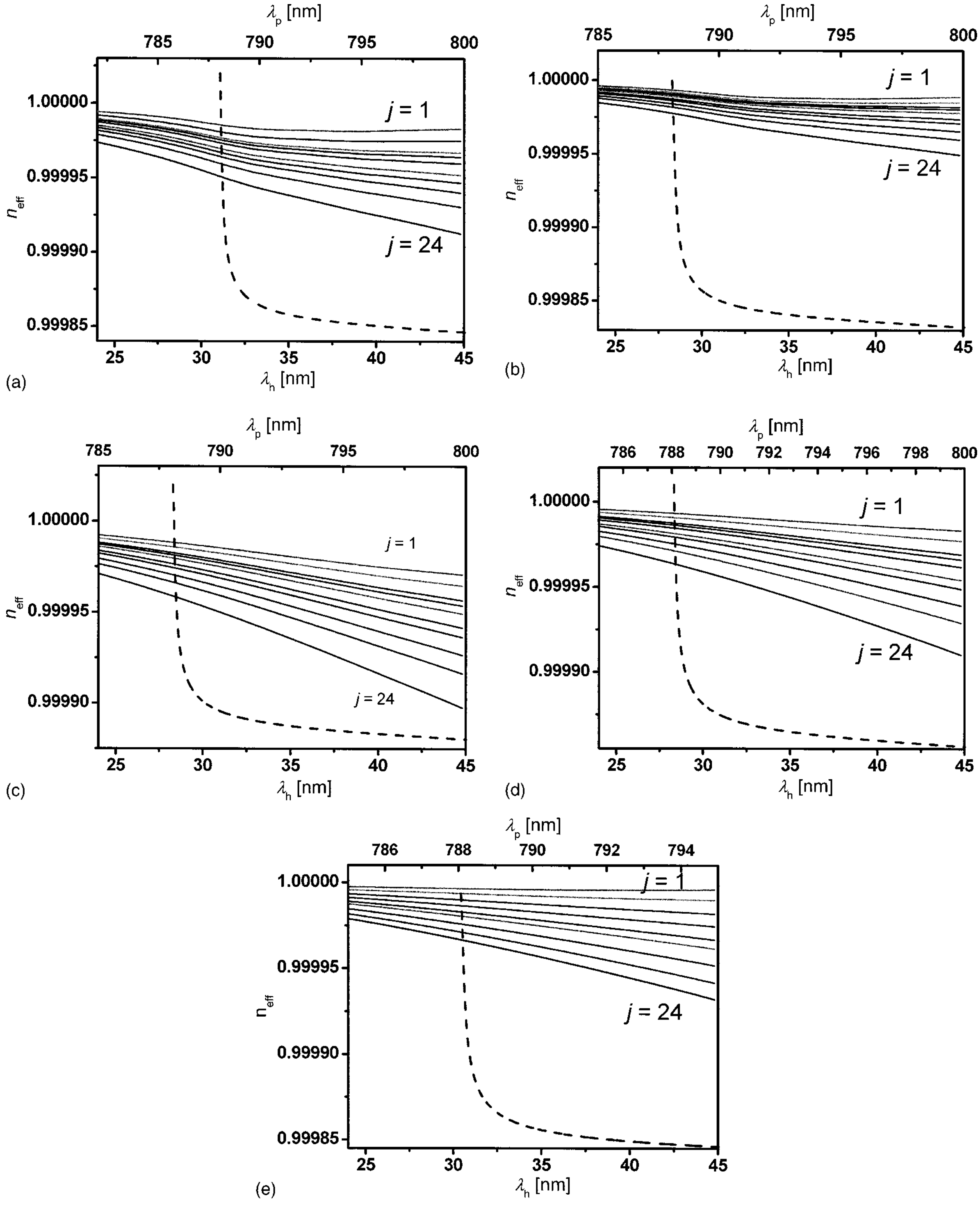

FIG. 6. Effective refractive indices for the fundamental mode of the pump field (the dashed line) and the fundamental ($j=1$) and higher order ($j=2-13,18,24$, from top to bottom) modes of high-order harmonics (with intensity profiles shown in Fig. 2) shown as functions of the pump and harmonic wavelengths (the upper and lower horizontal axes, respectively) for a hollow PCF with $\Lambda=10$ μm filled with (a, b) argon, (c, d) krypton, and (e) helium at a pressure of (a, c, e) 0.1 atm and (b, d) 0.05 atm.

or to switch the harmonic number corresponding to the maximum efficiency of frequency conversion.

The general conclusion that follows from the results presented in Figs. 3–6 is that the phase-matching strategies in the case of high-order harmonic generation in hollow-core PCFs may substantially differ from the recipes of phase-matched harmonic generation in standard, solid-cladding hollow fibers. Physically, this is due to a very strong waveguide dispersion at the frequency of the pump field characteristic of hollow PCFs with a small core diameter. Unlike standard hollow fibers, where the compensation of the material dispersion of the gas by the waveguide dispersion is at the heart of the phase-matching strategy [4–10], the waveguide component of dispersion for the pump field in hollow PCFs with a small core diameter far from the passband edges is typically much larger than the gas dispersion (the range of pump wavelengths from 795 to 810 nm in Figs. 3 and 4). However, closer to the edges of passbands, which are known to map PBGs of the PCF cladding, dispersion of guided modes bends, allowing phase matching to be achieved for high-order harmonic generation (the range of pump wavelengths from 787 to 790 nm in Figs. 3 and 4). The wavelength where the phase matching is achieved can be broadly tuned by choosing parameters of the hollow PCF [Figs. 4(a), 4(b), 5(a), and 5(b)] and varying the gas sort [Fig. 5(c)], the gas pressure [Figs. 3(a) and 3(b)], and the guided mode of harmonic radiation [Figs. 6(a)–6(e)]. The main drawbacks of this strategy are associated with radiation guiding losses, which noticeably increase near the passband edges and the limited bandwidth of the frequency range where the phase mismatch remains low. The former difficulty can be resolved by using hollow PCFs with low losses, recently demonstrated by Bouwmans *et al.* [18] and Smith *et al.* [20]. Radiation losses can be kept at the level of substantially less than 1 dB/m close to the edges of the passbands in these fibers, which is a tolerable level of waveguide losses for harmonic-generation experiments. The limited bandwidth of the phase-matching region, on the other hand, does not prevent efficient conversion of the pump energy to a single harmonic [the 21st harmonic of the 788-nm pump field in the case of Figs. 5(a) and 5(b) and the 19th harmonic of a pump field with the same wavelength in Fig. 5(c)], with fine adjustments of phase matching possible through gas-pressure and mode index variations. A radical broadening of the phase-matching range can be achieved by designing large-core hollow PCFs. Single-mode waveguiding can still be provided for the pump field in such fibers with properly engineered, sufficiently narrow PBGs of the PCF cladding.

The sensitivity of phase-matching conditions to the pressure of the gas filling the PCF core raises an interesting and practically important question as to whether absorption of pump and harmonic radiation can induce changes in the gas temperature sufficient to noticeably disturb the phase matching, leading to the requirement of temperature stabilization. Since the speed of sound is much lower than the speed of light, the absorbed pump and harmonic energy cannot be converted into noticeable gas-concentration variations within a single laser pulse, leading to no perturbations of the phase-matching condition. To examine gas-temperature changes induced by a sequence of pump pulses generating high-order harmonics in the gas filling the fiber core, we solve the one-dimensional heat conduction equation

$$\frac{\partial T}{\partial t} = \frac{\partial}{\partial x}\left(K(x,T,t)\frac{\partial T}{\partial x}\right) + f(T,t,x),$$

where T is the temperature, t is the time variable, x is the propagation coordinate, K is the temperature conductivity, and $f(T,t,x)=f(T,x,t)=2\Delta\varepsilon T(x)\delta(t-gN)/(3pS)$ for $L/2 \leqslant x \leqslant L/2$ and equal to zero otherwise is the function describing the heat source due to radiation energy absorption (g is the pulse repetition rate, N is the pulse number, $\Delta\varepsilon$ is the energy absorbed per pulse, S is the fiber core area, p is the gas pressure, δ is the delta function, and L is the fiber length). We consider 800-nm laser pulses with a pulse energy of 5 μJ and a pulse repetition rate of 1 kHz propagating through a hollow fiber with a length of 2 cm filled with argon at a pressure of 0.1 atm. The temperature conductivity is then $K \approx 1.7$ cm^2/s and absorption coefficients for the pump and harmonic radiation are 10^{-5} and 10 cm^{-1}, respectively. The maximum efficiency of pump energy conversion into the harmonic radiation is taken equal to 5×10^{-5}. To define boundary conditions for the considered heat conduction problem, we assume that the fiber is placed in a gas chamber with a sufficiently large volume, with the temperature of gas-chamber walls remaining constant. Absorption of pump and harmonic radiation energy in the hollow fiber then shifts the wavelength of exact phase matching for 21st-harmonic generation in argon [788.13 nm in Fig. 5(a)] by 0.03 nm, giving rise to a temperature-induced effective-index mismatch $(\delta n_{21})_T \approx 5\times 10^{-6}$. The influence of absorption-induced gas heating can, of course, increase in the case of ultracompact gas chambers with sizes close to the size of the hollow fiber used for harmonic generation. Temperature stabilization of gas-chamber walls would then help to compensate for the phase mismatch induced by the absorption of the pump and harmonic radiation energy.

V. CONCLUSION

Analysis performed in this work shows that hollow-core photonic-crystal fibers offer interesting phase-matching solutions for high-order harmonic generation by an isolated guided mode of pump radiation. We have demonstrated phase-matching regimes for the fundamental guided mode of pump field with a wavelength around 800 nm, generating harmonics within the wavelength range of 25–50 nm in hollow photonic-crystal fibers filled with argon, krypton, and helium. Geometric parameters of the fiber structure and the pressure of the gas filling the fiber core have been shown to serve as important, often orthogonal, control knobs, allowing a fine adjustment of the phase matching for high-order harmonic generation.

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