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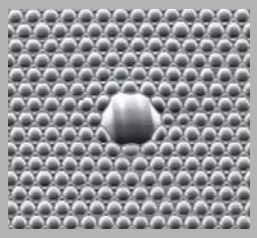
Abstract: We demonstrate single-mode guiding in a 4-μm-inner-diameter hollow photonic-crystal fiber filled with a highly nonlinear liquid. The nonlinear response of such a fiber is shown to drastically differ from a typical nonlinear response of a silica waveguide. Strong inertia of optical nonlinearity of the liquid filling the fiber core translates into a pulse-width-dependent red shift of the spectrally broadened fiber output.



Benzene-filled PCF structure

Understanding the nonlinear-optical response of a liquid-core photonic-crystal fiber

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Liquid-core waveguide structures have long been known and intensely used in nonlinear optics. In particular, capillary waveguides filled with highly nonlinear liquids have been employed in pioneering experiments on self-phase modulation [1] and nonlinear Raman scattering [2]. In a more recent work, an interest in waveguides filled with liquid-phase materials has been twofold. Firstly, many liquid materials are known to exhibit strong optical nonlinearity [3]. Filling waveguides with such materials offers much promise for a radical enhancement of guided-wave nonlinear-optical processes [4–6].

Secondly, optical nonlinearity of liquid-phase materials is known to originate from different types of rather complicated molecular motions [7]. Many fundamental questions concerning the relation between the molecular dynamics and optical nonlinearity of liquid materials still remain open [8]. Waveguide structures offer a convenient platform for the experimental investigation into these fundamental aspects of the nonlinear optics in the liquid phase on different time scales [9,10].

Photonic-crystal fiber (PCF) technologies [11,12] enhance performance and offer new functionalities of liquid-filled waveguides [13] as tools for nonlinear optics. S. Yiou et al. [14] have recently demonstrated efficient stimulated Raman scattering in a hollow PCF filled with ethanol. A. Bozolan et al. [15] have reported supercontinuum generation in a water-core PCF pumped near the zero-dispersion wavelength of water. Dispersion tailoring in liquid-containing PCFs and PCF-like waveguides, including those designed for efficient nonlinear-optical transformation of laser pulses, has been theoretically analyzed in [16,17].

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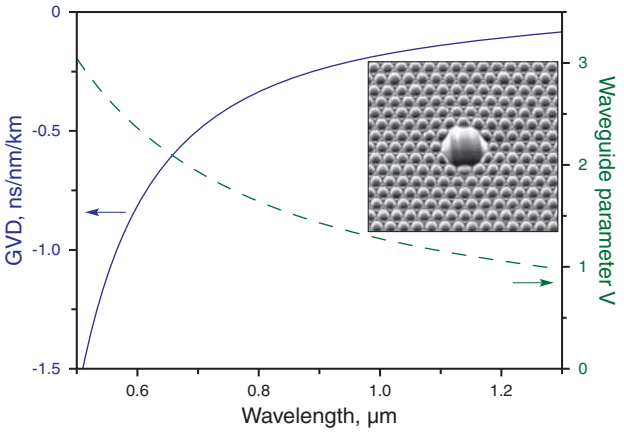

Figure 1 (online color at www.lphys.org) The group-velocity dispersion (GVD, solid line) and waveguide parameter V (dashed line) calculated as a function of the radiation wavelength for a benzene-filled PCF structure shown in the inset. The fiber core diameter is 4 μm

In this paper, we demonstrate a hollow photonic-crystal fiber (PCF) that supports single-mode guiding at wavelengths longer than 600 nm in a 4-μm-diameter liquid-filled core, thus offering an attractive platform for nonlinear-optical experiments in the liquid phase. This PCF is employed to demonstrate that liquid-phase materials can radically modify the nonlinear-optical response of a waveguide structure relative to a typical nonlinear response of a silica waveguide. We show that the strong inertia of optical nonlinearity, characteristic of highly nonlinear liquid-phase materials, gives rise to a pulse-width-dependent spectral red shift of the spectrally broadened fiber output. This wavelength shift remains strong even for pulse widths as large as several hundreds of femtoseconds.

The photonic-crystal fiber used in this work was produced by means of a standard PCF stack-and-draw technique [11,12]. The hollow core of this fiber has a diameter of about 4 μm and is surrounded by a dual cladding (the inset in Fig. 1), consisting of an inner, microstructured part with a thickness of about 22 μm and an outer, solid silica cladding section. When filled with liquid-phase benzene, such a structure can serve an index-guiding fiber [18], with the refractive index of the liquid core exceeding the effective index of the microstructured, silica–benzene part of the fiber cladding.

For a qualitative understanding of the guiding properties of our benzene-filled liquid-core PCF, we use a standard definition of the waveguide parameter

$$V = \frac{2\pi a}{\lambda}\sqrt{n_{core}^2 - n_{clad}^2}\,,$$

where a is the core radius, λ is the radiation wavelength, n_{core} is the refractive index of the core, and n_{clad} is the effective refractive index of the microstructure composite cladding. The dashed line in Fig. 1 presents the parameter V for the studied liquid-core PCF as a function of radiation wavelength with the effective refractive index of the cladding n_{clad} calculated by using the finite-element method (FEM). Material dispersion is included in these calculations through the Sellmeier equation for silica [19] and the Cauchy formula for benzene at 20°C [20]. As can be seen from Fig. 1, the condition of single-mode guiding, $V < 2.405$ is satisfied for $\lambda > 590$ nm. The FEM calculations also show that the benzene-filled PCF with a spatial structure shown in the inset to Fig. 1 supports single-mode guiding at least for $\lambda > 600$ nm and provides normal dispersion everywhere within the range of wavelengths studied in our experiments. The wavelength dependence of the group-velocity dispersion (GVD) for this fiber is shown by the solid line in Fig. 1.

The laser system used in our experiments was based on a Cr^{4+}:Forsterite master oscillator [21] pumped with an ytterbium fiber laser. It generated 30 – 60-fs light pulses of radiation with a central wavelength of 1.25 μm at a repetition rate of 120 MHz. These pulses were transmitted through a stretcher and an isolator, to be amplified in a Nd:YLF-laser-pumped amplifier and recompressed to a variable pulse width of 100 – 800 fs with a pulse energy up to 50 μJ at 1 kHz. In the experiments, the second-harmonic output of the Cr:Forsterite laser with a central wavelength of 619 nm and a pulse width of 200 fs was used to study nonlinear-optical effects in a liquid-core PCF.

To understand the nonlinear-optical transformation of light pulses in a benzene-filled PCF, we adopt a standard approach based on the use of the time-domain response function of a nonlinear material, $H(\theta)$, which is conventionally defined through the intensity-dependent nonlinear phase shift of a light field as [3,22]

$$\Phi_{nl}(\eta) = \gamma z \int_{-\infty}^{\eta} H(\eta-\theta)\left|A(\theta)\right|^2 d\theta\,, \qquad (1)$$

where η is the retarded time, z is the propagation coordinate, $A(\eta)$ is the temporal envelope of the light field, $\gamma = 2\pi\lambda^{-1}n_2 S_{eff}^{-1}$ is the nonlinear coefficient, n_2 is the nonlinear refractive index, and S_{eff} is the effective mode area. The response function

$$H(\theta) = (1-f)\delta(\theta) + f\Theta(\theta)R(\theta) \qquad (2)$$

includes an ultrafast Kerr-type term involving the delta function $\delta(\theta)$, which mimics a nearly instantaneous electron response, and an inertial term with the Heaviside step function $\Theta(\theta)$ and the retarded response function $R(\theta)$, which describes a complex manifold of slower molecular motions in the liquid. The weighting factor f in Eq. (2) defines the fractional contribution of the retarded term to the overall nonlinear response. Since the temporal width of the laser pulses in our experiments (about 200 fs) was chosen in such a way as to be long enough to avoid excitation of the most intense Raman-active vibrations of benzene

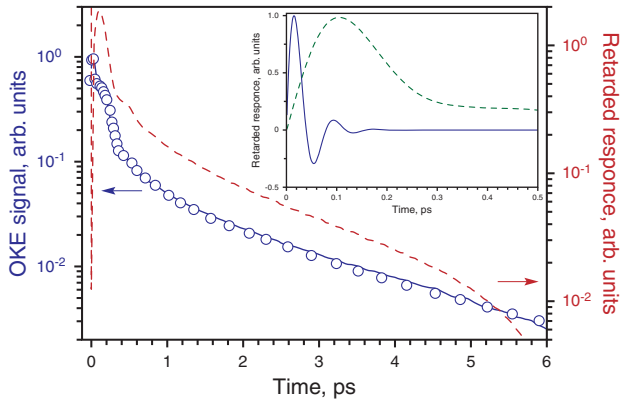

Figure 2 (online color at www.lphys.org) An OKE signal measured by M. Ricci et al. [23] as a function of the delay time between the pump and probe pulses (open circles) and the best fit for this dependence (solid line) achieved with the time-domain nonlinear response function $H(\theta)$ with $f = 0.76$ (shown by the dashed line). The inset shows the damped-oscillator Raman response function of fused silica (solid line) and the retarded response $R(\theta)$ related to librations in benzene (dashed line)

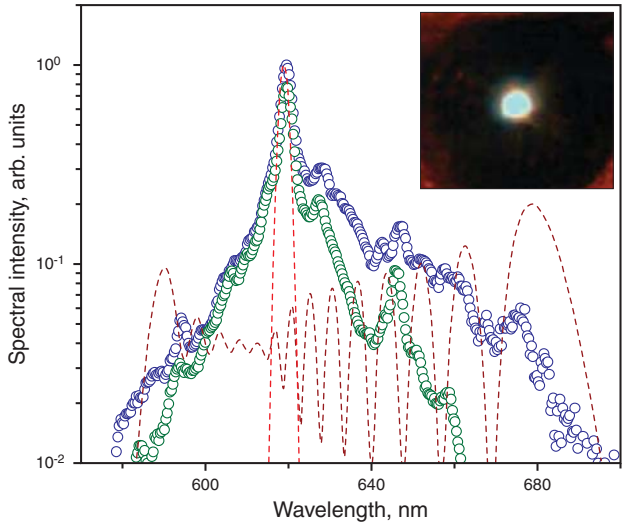

Figure 3 (online color at www.lphys.org) The spectrum of second-harmonic pulses of a Cr:Forsterite laser transmitted through the benzene-filled PCF. The input pulse width is 200 fs. The input pulse energy is 4 nJ (open circles) and 6 nJ (filled circles). The spectrum of the input light pulse is presented by the dashed line. The dotted line shows the spectrum of a light pulse with an initial pulse width of 200 fs and a nonlinear phase shift as defined by Eq. (1) with $f = 0.76$ and the retarded response $R(\theta)$ as shown in the inset to Fig. 2. An image of the PCF output end is shown in the inset

(including the 992-cm^{-1} symmetric stretch mode), the retarded part of the optical nonlinearity of benzene in our experiments is mainly due to librational motions of benzene molecules.

To find the retarded response $R(\theta)$ in Eq. (2), we take a Fourier transform of typical optical Kerr effect (OKE) spectral data for librations in benzene [23]. The $R(\theta)$ function generated with the use of this procedure (shown by the dashed line in Fig. 2) radically differs from the standard damped-oscillator model of the Raman response of fused silica (cf. the inset in Fig. 2). The behavior of the $R(\theta)$ function of benzene is generally much more inertial. On the picosecond time scale, this function, as can be seen from Fig. 2, exhibits a purely exponential decay, related to the orientational relaxation of benzene molecules [23]. To find the fractions of the ultrafast electronic and inertial molecular nonlinearities in the response function $H(\theta)$, we use Eq. (1) and Eq. (2) to fit the time-resolved OKE signal measured by M. Ricci et al. [23] (shown by open circles in Fig. 2) as a convolution of $H(\theta)$ and a Gaussian probe pulse with a pulse width of 50 fs (modeling light pulses employed in experiments of [21]) using f as a fitting parameter. The best fit (the solid line in Fig. 2) is achieved with $f = 0.76$.

For a benzene-core PCF with a structure as shown in the inset to Fig. 1, the FEM analysis gives $S_{eff} \approx 15\ \mu\text{m}^2$. With a typical estimate for the nonlinear refractive index of benzene $(n_2)_b \approx 2\times10^{-15}$ cm^{2}W^{-1}, this yields $\gamma \approx 40$ W^{-1}km^{-1}. Since the n_2 of benzene is substantially higher than the nonlinear refractive index of silica ($(n_2)_s \approx 3\times10^{-16}$ cm^{2}W^{-1}), the nonlinear coefficient γ for a benzene-core fiber is much larger than the γ value for a silica waveguide with the same effective mode area. However, the fractional contribution of the ultrafast electron nonlinearity to the nonlinear response of a benzene-core fiber is much lower ($1 - f \approx 0.24$) than that for a silica fiber, where the $1 - f$ fraction is as high as 0.82. The much stronger inertia of the nonlinear response of a benzene-core fiber translates into a much more pronounced red shift of laser pulses transmitted through the fiber, and into a much stronger sensitivity of the spectral broadening of these pulses to the pulse width.

In Fig. 3, we present experimental results that illustrate the key tendencies in the nonlinear-optical transformation of light pulses in our liquid-core fiber. In these experiments, the 200-fs, 619-nm second-harmonic output pulses of the Cr:Forsterite laser were launched into a 10-cm piece of benzene-filled hollow PCF with the cross section structure shown in the inset to Fig. 1. The spectrum of the input laser pulses is shown by the dashed line in Fig. 3. Predominantly single-mode guidance of light within the studied range of wavelengths is verified by the beam profile of the fiber output (the inset in Fig. 3). Light pulses with a higher input energy experience a stronger spectral broadening and a more pronounced red shift (cf. open- and filled-circle curves in Fig. 3).

To provide a qualitative insight into the results of these measurements, we also plot in Fig. 3 the spectrum of a light field

$$E(\eta) = A(\eta) \exp\left[i\omega_0\eta + i\Phi_{nl}(\eta)\right],$$

where ω_0 is the central frequency of the input field and the nonlinear phase shift $\Phi_{nl}(\eta)$ is defined by Eq. (1) with the nonlinear response $H(\theta)$ of benzene taken with $f = 0.76$ and the retarded response $R(\theta)$ as shown in the inset to Fig. 2. With $\gamma = 40$ W^{-1}km^{-1}, an initial pulse energy of 6 nJ, an input pulse width of 200 fs, and a fiber length of 10 cm, this simple model, as can be seen from Fig. 3, not only reproduces the main tendencies in the red shifting and spectral broadening of the light pulses transmitted through the liquid-core PCF, but also provides a reasonable prediction for the bandwidth of the spectrally broadened pulses, as well as for the high- and low-frequency edges of the output spectra. Although there is no way that this simple model could reproduce also the shape of the broadened spectra at the output of the fiber, deviations of the experimental spectra from the results of calculations in Fig. 3 are still quite instructive. These deviations are especially noticeable around the carrier frequency of the input field, indicating that, because of a strong scattering inside the liquid core and on the input end of the fiber, a substantial fraction of radiation energy is transported through the fiber outside its highly nonlinear liquid core. This radiation is readily seen in an image of the output end of the fiber.

We have demonstrated in this work that a hollow PCF filled with a highly nonlinear liquid can support single-mode guiding at wavelengths longer than 600 nm in a 4-μm-diameter liquid core. The nonlinear response of such a fiber has been shown to drastically differ from a typical nonlinear response of a silica waveguide. The strong inertia of optical nonlinearity of the liquid filling the fiber core translates into a pulse-width-dependent red shift of the spectrally broadened fiber output. Liquid-core fibers with carefully controlled dispersion and nonlinearity are envisaged to enhance the potential of fiber-based laser sources, frequency converters, and optical field waveform synthesizers [24–28].

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