Extreme optical nonlinearities in chalcogenide glass fibers embedded with metallic and semiconductor nanowires

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A nanostructured chalcogenide-metal optical fiber is proposed. This hybrid nanofiber is embedded with a periodic array of triangular-shaped deep-subwavelength metallic (or semiconductor) nanowires set up in a bowtie configuration. Calculations show that the proposed nanostructured fiber supports a guided collective plasmonic mode enabling both subwavelength field confinement and extreme nonlinear light-matter interactions. A scheme is also proposed whereby the large linear absorption losses are compensated via nonlinear optical gain when semiconducting single-wall carbon nanotubes are used in place of metal.

In the last decade, photonic crystal fibers and microstructured optical fibers have demonstrated exceptional control over the group velocity chromatic dispersion. Recent structured optical fibers have demonstrated exceptional interaction lengths inside nonlinear media. In parallel, the advances in micro- and nano-fabrication techniques have sparked thriving research on microstructured optical fibers with subwavelength features and using high refractive index compound glasses. These so-called emerging waveguides allow the exploration of new operation regimes where tight field confinement, enhanced light-matter nonlinear interactions and dispersion engineering combine to enable long interaction lengths inside nonlinear media. In parallel, the merging of plasmonics with integrated optics has shown vast potential for sensing and the transmission and modulation of optical signals on the subwavelength scale. Light guiding mediated by metallic nanowire arrays in optical fibers was also investigated and their potential for nonlinear optical plasmonics was mentioned in Ref. 9 but not studied in detail.

In this letter, we present a new type of nonlinear metallo-dielectric nanostructured optical fiber (NOF): the chalcogenide fiber with deep-subwavelength metallic inclusions. We demonstrate that the extreme field intensities obtained at the sharp edges of the subwavelength metallic nanowires enable giant nonlinear optical enhancements to be achieved. Moreover, we show that the large propagation losses due to Ohmic absorption in the metal may be compensated by replacing the noble metal filling with a semiconducting fiber exhibiting two-photon gain (TPG).

When fabricating a fiber comprising several dielectric rods (or capillaries) using the stack-and-draw procedure, the empty interstitial holes [Fig. 1(a)] that may occur between adjacent rods are usually treated as unwanted defects. Here, we fill these nanovoids with metal (or a semiconductor) such that we obtain a symmetrical periodic array of triangle-shaped metallic nanowires. Coincidentally, the triangular “bowtie” structure is known to be a very effective configuration for yielding strong field confinement in the nanogap between two apices. In the proposed fiber design, the intense local fields enabled by metallic bowtie nanowires [Fig. 1(b)] enhance the nonlinear light-matter interaction within the chalcogenide glass matrix.

Towards the fabrication of metal, or semiconductor, nanowires embedded in a dielectric matrix, a practical method was recently demonstrated in Ref. 14 where the authors used a consecutive stack-and-draw technique to produce nanowires with extreme aspect ratios. A potential fabrication strategy of this NOF is to first stack identical circular glass rods in a triangular lattice configuration in the preform and then allow the rods to overlap during the thermal softening and drawing process. The ensuing small air interstices are then filled by pumping molten metal at high pressure, as demonstrated in Ref. 9. This interstice-filling approach allows the desirable edge-to-edge alignment of pairs of nanowires to be enforced right from the initial macroscopic preform.

For the first hybrid fiber studied, the wavelength of interest is \( \lambda = 3.0 \) \( \mu m \) and the chosen noble metal is gold (Au) while the dielectric material is AsSe3 chalcogenide glass which has a wide transparency inside the middle-infrared (2–12 \( \mu m \)). Both materials present large values of the nonlinear index: \( n_{2}^{Au} = 1.32 \times 10^{-12} \) \( m^2/W \) and \( n_{2}^{AsSe} = 1.1 \times 10^{-17} \) \( m^2/W \).

![FIG. 1. (Color online) (a) \( S_z \)-flux distribution of the fundamental plasmonic supermode in the chalco-gold nanostructured fiber \( (r_{fiber} = 0.450 \) \( \mu m \), \( f_{3} = 0.03 \)) at \( \lambda = 3.0 \) \( \mu m \) and (b) close-up view of the enhanced local fields (arrows denote vector \( E \)-fields) in a “bowtie” pair of nanowires. Inset (a): close-up view of the geometrical configuration of overlapping rods and interstices.](image-url)
We also investigated hybrid fibers filled with semiconducting single-wall carbon nanotubes (SWCNs) instead of gold. A closely packed ensemble of SWCNs having a Gaussian distribution of average radius $R_{avg} = 0.67$ nm and standard deviation $R_{std} = 0.05$ nm was assumed. When excited by light polarized along their symmetry axis, such SWCNs are expected to behave as a metal ($\sigma < 0$) in the near-infrared ($\lambda = 1.35 - 1.85 \mu m$) and, crucially, provide TPG ($\chi^2 = -3.42 \times 10^{-8}$ m/W) and a large nonlinear index ($n_2^{SWCN} = 1.90 \times 10^{-15}$ m$^2$/W) at the pump wavelength $\lambda = 1.5 \mu m$.\(^1\)

To simplify analysis, the present investigation focuses on the case of $N = 2$ layers of rods (of equal radii $r_{rod}$) with gold-filled interstices [Figs. 1(a) and 1(b)]. In our model, the overlapping of rods is tuned by the “overlap half-distance” $\delta$ as defined in Fig. 1(b), and in practice this is accomplished by controlling the injected gas pressure when drawing the fiber. Here, we use the convenient non-dimensional parameter $f_\delta = \delta/r_{rod}$ where $f_\delta = 0$ defines the case of tangent circles; while positive values $0 < f_\delta < f_{\delta,max}$ controls the overlapping of adjacent rods up to a maximal ratio $f_{\delta,max} = 0.1339$ (i.e., the limit where the area of the triangular-shaped interstice disappears). The total outer radius of the fiber is set as $r_{fiber} = 5r_{rod} - 3\delta$ such that the whole geometry can be specified using only $r_{fiber}$ and the “overlap factor” ($f_\delta$) which was kept fixed at 0.03 in this study, corresponding to 2.8% by volume of metallic content.

Recently, there have been demonstrations of large nonlinear optical enhancement in chalcogenide nanowires.\(^1\) Therefore, as a benchmark, we compare below the optical properties of our hybrid NOF with the bare nanowire (i.e., rod-in-air). Using fully vectorial finite-element simulations, we solved for the fundamental HE$_{11}$ mode guided in the bare As$_2$Se$_3$ chalcogenide nanowire and for the fundamental plasmonic supermode [Fig. 1(a)] guided in the metallo-dielectric NOF, for various fiber radii.

The effective refractive index $Re(n_{eff})$ and linear absorption losses ($x_2$) of the fundamental guided mode in the chalcogenide nanowire and in the two studied hybrid NOFs as a function of fiber radius are plotted in Fig. 2(a) and Fig. 2(b), respectively. As shown in the inset of Fig. 2(a), the fraction of guided power of the fundamental mode in the bare nanowire rapidly becomes almost 100% confined inside the chalcogenide glass matrix (and virtually zero in the air cladding) as the radius of the fiber increases. Consequently, we observe in Fig. 2(a) for the nanowire $n_{eff} \rightarrow n_{bulk}$ as $r_{fiber} \gg \lambda$, where $n_{bulk} = 2.789$ at $\lambda = 3.0 \mu m$ for As$_2$Se$_3$ chalcogenide glass. The reverse is also true in the limit of weak guidance: the value of $n_{eff}$ reaches the refractive index of air cladding such that $n_{eff} \rightarrow n_{air} = 1$ as $r_{fiber} \ll \lambda$. On the other hand, for the chalco-gold NOF, the value of $n_{eff}$ increases with reduction of the fiber radius. This behavior can be rationalized by considering the fraction of power that propagates inside metallic regions [inset of Fig. 2(a)]. We first note that the geometrical proportions of the fiber’s internal structure are preserved as the radius gets smaller. Thus for very small NOF radii ($r_{fiber} < 1 \mu m$), the dimensions of the metallic nanowires become deeply subwavelength and comparable to the skin depth in metal such that a significant fraction of guided power overlaps with the strongly dispersive metallic regions. Due to the semiconducting and absorbing nature of SWCNs, the corresponding $n_{eff}$ curve is located between that of a noble metal (gold)-based NOF and that of a pure dielectric nanowire.

The calculations also indicate that the proposed hybrid NOFs can theoretically yield nonlinearities over $1 \times 10^4$ W$^{-1}$m$^{-1}$ [Fig. 3(a)]. In particular, the nonlinearity reaches a peak of $\gamma = 286$ W$^{-1}$m$^{-1}$ at $r_{fiber} = 0.415 \mu m$ for the chalco nanowire; while the chalco-SWCNs and the chalco-gold NOFs yield, respectively, $\gamma = 2.04 \times 10^4$ W$^{-1}$m$^{-1}$ and $\gamma = 1.34 \times 10^5$ W$^{-1}$m$^{-1}$, corresponding to more than 2 orders of magnitude increase in nonlinearity over a bare nanowire of the same size. The inset of Fig. 3(a) plots the effective mode area ($A_{eff} = \int S_z dA/\int S_z dA$) and indicates that hybrid fibers consistently provide better field confinement over their simple nanowire counterparts of the same size. Specifically, for $r_{fiber} < 0.415 \mu m$, while the effective mode area of a chalco nanowire diverges to the completely unguided limit ($A_{eff} \rightarrow \infty$ for $r_{fiber} \ll \lambda$), the mode area supported by a hybrid nanofiber keeps decreasing with smaller fiber radii. This nanoscale localization of light beyond the diffraction limit is made possible by the plasmonic guiding in the metallic (or semiconductor) nanowire arrays.

The nonlinear optical enhancement factor in chalcometallic fibers (compared to similar nanowires) is consistently greater than 20 across the investigated range of fiber radii. The nonlinear parameter was computed using the vectorial definition\(^1\)
The group velocity chromatic dispersion (GVD) was defined as \( v/c \) total \( \approx \) constant (TPA regime). For our given set of (average, radial) values, the SWCNs are excited in the TPG regime (i.e., \( \Delta_{2} < 0 \)) at \( \lambda = 1.5 \mu \text{m} \). The intensity threshold for the compensation of linear losses by the nonlinear gain is found by setting Eq. (2) to zero: \( \Delta_{2} = -\Delta_{1}/(\Gamma_{2}) \). Moreover, a pulse peak power threshold is defined as \( P_{T} = P_{T_{\text{th}}} \) which specifies the minimum power of the incident pulse required in order to compensate lossless modal propagation. Figure 3(b) indicates that peak powers as low as a few hundred watts (for small fiber radii) would be necessary to accomplish this.

The group velocity chromatic dispersion (GVD) was computed using the definition \( D = -\lambda/c(d^{2}n_{\text{eff}}/d\lambda^{2}) \). Figure 4 shows that via appropriate choice of the geometrical parameters, the chalcogen-metallic NOF enables engineering of the GVD to create a zero-dispersion point (ZDP) at \( \lambda = 3.0 \mu \text{m} \) operation wavelength (Fig. 4(a) or \( \lambda = 1.5 \mu \text{m} \)) Fig. 4(b)). We also note some regions where the GVD is engineered to be slightly anomalous, thus allowing nonlinear solitonic effects to occur. Moreover, the ultrafast response (< 50 fs) of the third-order nonlinearity in chalcogenide-based waveguides indicates that the proposed NOF can potentially achieve broader bandwidths than all-optical processing devices operating on a resonant-based nonlinearity and without free-carrier effects (in the case of gold) that are present in other materials such as silicon. Our simulations also point to large group indices, defined as \( n_{g} = n_{\text{eff}} + \omega(dn_{\text{eff}}/d\omega) \), with values > 20 which could be achieved concurrently in the hybrid fibers. Therefore, this result suggests that the proposed NOF design is not only useful for extreme nonlinear optical interactions but also for slow light applications with an engineered chromatic dispersion profile.

In conclusion, a novel type of NOF has been proposed. Our calculations demonstrate that the hybrid chalcogenide-metallic NOF provides a platform for achieving nanoscale mode area nonlinear light-matter interactions and also constitutes an alternative path for investigating slow-light applications. Moreover, we demonstrate that substitution of the noble metal content for semiconducting single-wall carbon nanotubes opens the way for the compensation of losses through nonlinear optical gain. Finally, we emphasize that the proposed chalcogen-metallic fibers are relevant to the study of extreme nonlinear light-matter interactions, all-optical signal processing, and low-powered highly integrated nonphotonics devices in general.

\[ dI/dz = -\lambda I - \Gamma_{2} I^{2}, \quad (2) \]

where \( \lambda_{1} \) and \( \lambda_{2} \) denote, respectively, the linear and nonlinear absorption coefficients, and \( \Gamma_{2} \) is the modal confinement factor inside the active medium [see inset of Fig. 2(a)]. Values of \( \lambda_{2} > 0 \) correspond to the two-photon absorption (TPA) regime. For our given set of \( (R_{\text{avg}}, R_{\text{al}}) \) values, the SWCNs are excited in the TPG regime (i.e., \( \lambda_{2} < 0 \)) at \( \lambda = 1.5 \mu \text{m} \). The intensity threshold for the compensation of linear losses by the nonlinear gain is found by setting Eq. (2) to zero: \( \lambda_{2} = -\lambda_{1}/(\Gamma_{2}) \). Moreover, a pulse peak power threshold is defined as \( P_{T} = P_{T_{\text{th}}} \) which specifies the minimum power of the incident pulse required in order to compensate lossless modal propagation. Figure 3(b) indicates that peak powers as low as a few hundred watts (for small fiber radii) would be necessary to accomplish this.