Robust plasmonic tips fabricated by the tapering of composite hybrid silicate microfibers with metallic core

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ABSTRACT

The development of plasmonic devices for sensing applications can offer high sensitivity and a dramatic improvement to the detection limits due to the high field enhancement at the metal surfaces. The platform proposed here is a tapered hybrid microfiber comprising a metal core and a glass cladding. The existence of a glass cladding not only serves as a mechanical host for the metal core, but also provides ease of handling regarding the tapering process. The advantages of this composite material system over pure metal tips are the absence of impurities and the multiple excitation of the plasmon modes due to the total internal reflection at the glass/air interface. The improved field enhancement at the apex of these tapered microwires was calculated through Finite Element Method (FEM) simulations. Enhancement factors up to 10^4 were theoretically observed for this type of tapered microwires. The use of different metals having different melting points and thermal expansion coefficients as well as different glass thicknesses can lead to an optimization of the tapering process conditions in order to achieve tapered microwires with the desirable geometrical characteristics.

Keywords: Plasmonics, localized surface plasmon resonance, hybrid microfibers, metals, fiber tapers

1. INTRODUCTION

Surface plasmon polaritons (SPPs) are surface electromagnetic waves travelling along a metal/dielectric interface originated from coupling between oscillating electrons and incident photons. SPPs and localized surface Plasmon resonances (LSPR) can offer high field enhancement and confinement beyond the diffraction limit and have found many applications in biosensing, surface-enhanced Raman spectroscopy (SERS) and also microscopy.

In this paper a tapered hybrid gold-core/borosilicate-cladding microfiber is studied. Their advantages over the metal tips are the ease of light coupling and the re-excitation of the plasmon modes due to the total internal reflection at the dielectric/air interface through the length of the fiber. Although the field enhancement of pure metal tips has been extensively studied theoretically¹⁻³, it has been clearly identified lately the need for further research⁴ towards the engineering of physically realizable and robust plasmonic structures with enhanced performance. The absence of impurities and the multiple excitation of the nanowire due to the repeated silicate glass/air total internal reflections⁵ can provide high field enhancement at the apex of such tapers by overcoming the metal induced losses. For the tapering the method of heating and stretching is used for the accurate control of the final taper geometry.

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2. FABRICATION OF DIELECTRIC CLADDING/METAL CORE MICROFIBERS

The hybrid gold core/borosilicate cladding are fabricated in the Optoelectronics Research Center (ORC, University of Southampton) and have core diameters varying from 3-20 μ m. For the fabrication of the hybrid metal-core/glass-cladding microfibers a suitable metal should be used having melting point below the softening point of the glass cladding. The metals that were used were silver, copper, gold and aluminum having melting points ranging from 660 °C for aluminum to 1085 °C for copper (Fig. 1). For the glass cladding Schott Duran borosilicate tubing of different outer and inner diameters (81 wt% SiO₂, 13 wt% B₂O₂, 4 wt% Na₂O+K₂O, 2 wt% Al₂O₃) was identified as a promising ,widely available and low cost solution and therefore it was used⁶.

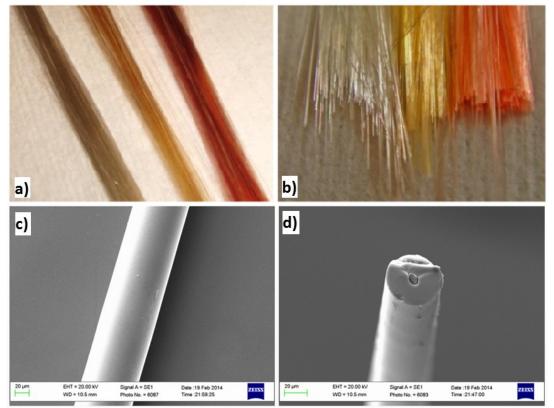


Figure 1. (a), (b) Bundles of silver, gold and copper-core borosilicate microfiber, (c), (d) SEM images of metal-core borosilicate microfibers

The microwires' fabrication process was proved to be not a trivial process and certain modifications in the "stack and draw" fiber drawing systems were needed. The main parameters that had to be controlled during the drawing process of the fibers are the temperature, feed rate, pull rate and volume of metal used together with a number of secondary parameters. The initial trials were based on copper based microwires due to the copper's low cost. Many fiber snaps occurred during initial copper-core fiber draws mostly due to the copper not melting fully, or too much copper flowing into the preform neck at once. For continuous core microfibers a Schott Duran borosilicate preform of 6 mm outer diameter and 2.5 mm inner diameter is fused at one end to create a seal, and four lengths of 0.5 mm diameter copper wire are inserted into the preform. No gas purge is supplied to the center of the furnace, where the preform is situated, but instead a nitrogen gas flow is delivered through the cooling ring. A number of carefully considered parameters in drum's speed were needed to be considered in order to achieve the continuous production of microwires at speeds of several tens of meters per minute, and of a solitary spool up to 50 km in length of continuous fiber microwire. However it was identified that for some metals there were metal core discontinuities in the entire spool length and therefore there is

currently under way an optimization process for production of continuous microwires in such long lengths. In plasmonic applications though, continuity over long length is not a crucial factor as small pieces of microwires are needed.

The core discontinuity that was apparent a number of studied microwires were further investigated. It was confirmed that in core discontinuous fibers, the mode profile at the output of these fibers is well preserved due to the continuous reexcitation effects into the silicate enclosure, through the length of the fiber as a result of repeated total internal reflection at the silicate/air interface⁵. We obtained the beam profile of a microfiber of 54µm diameter and 11µm core diameter. Figure 2 shows the beam profiles of a continuous and a discontinuous piece of the microfiber (see inset picture). The nanowires are excited by coupling light of 532nm into the borosilicate enclosure. As expected two peaks are observed in the 2D beam profile at the gold/glass interface corresponding to the illustrated modelled TM_{01} mode profile (inset picture). In order to obtain the beam profile we used a 60x objective lens to image the near-field intensity distribution at the fiber output and a scanning slit optical beam profiler.

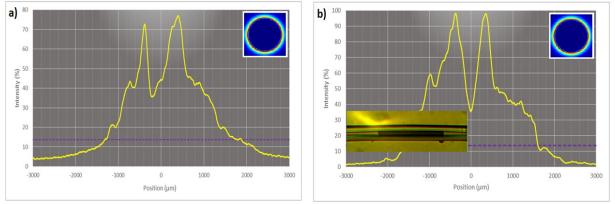


Figure 2. Mode profile of a hybrid microfiber with continuous (a) and discontinuous (b) core. The inset pictures show the TM_{01} mode profiles and a characteristic segment of a core discontinuous fiber.

3. MODELLING

For metallic core nano-wires of radius R_1 surrounded by a dielectric layer of radius R_2 , the amplitudes of the electric and magnetic fields for TM_{01} mode⁶ are:

$$E(\rho) = \begin{cases} A_1 \left[-i\left(\frac{\beta}{\alpha_1}\right) I_1(\alpha_1 \rho) \hat{e}_{\rho} + I_0(\alpha_1 \rho) \hat{e}_z \right], & \rho \le R_1 \\ \left\{ \left(\frac{\beta}{\alpha_2}\right) \left[-iA_2 I_1(\alpha_2 \rho) + B_2 K_1(\alpha_2 \rho) \right] \hat{e}_{\rho} + \left[A_2 I_0(\alpha_2 \rho) - iB_2 K_0(\alpha_2 \rho) \right] \hat{e}_z, & R_1 \le \rho \le R_2 \\ A_3 \left[\left(\frac{\beta}{\alpha_3}\right) K_1(\alpha_3 \rho) \hat{e}_{\rho} - iK_0(\alpha_3 \rho) \hat{e}_z \right], & \rho \ge R_2 \end{cases} \\ H_{\varphi}(\rho) = k \times \begin{cases} A_1 \left[-i(\frac{\varepsilon_1}{\alpha_1}) I_1(\alpha_1 \rho) \right], & \rho \le R_1 \\ \left(\frac{\varepsilon_2}{\alpha_2}\right) \left[-iA_2 I_1(\alpha_2 \rho) + B_2 K_1(\alpha_2 \rho) \right], & R_1 \le \rho \le R_2 \\ A_3 \left[\left(\frac{\varepsilon_3}{\alpha_3}\right) K_1(\alpha_3 \rho) \right], & \rho \ge R_2 \end{cases} \end{cases}$$

where β is the propagation constant, ε_1 , ε_2 and ε_3 are the relative permittivities of metal core, dielectric cladding and air respectively, $\alpha_j = (\beta^2 - \varepsilon_j k^2)^{1/2}$ for j=1,2,3, I and K are the modified Bessel functions of the first and second kinds and A_1 , A_2 and A_3 are the complex constants.

The dispersion equation can be derived by the continuity of the tangential components of the fields at the interfaces $\rho = R_1$ and $\rho = R_2$:

$$\varepsilon_{1}\alpha_{2}I_{1}(\alpha_{1}R_{1})[\alpha_{2}\varepsilon_{3}K_{1}(\alpha_{3}R_{2})M_{00} + \alpha_{3}\varepsilon_{2}K_{0}(\alpha_{3}R_{2})M_{10}] = -\varepsilon_{2}\alpha_{1}I_{0}(\alpha_{1}R_{1})[\alpha_{2}\varepsilon_{3}K_{1}(\alpha_{3}R_{2})M_{01} + \alpha_{3}\varepsilon_{2}K_{0}(\alpha_{3}R_{2})M_{11}]$$

where $M_{ab} = I_a(a_2R_2)K_b(a_2R_1) - (-1)^{a+b}I_b(a_2R_1)K_a(a_2R_2)$ with $a, b = \{0 \text{ or } 1\}$

Figures 3 show the real and imaginary parts of the effective refractive index (n_{eff}) as a function of the wavelength for different radii of the gold core. As the radius of the copper tip decreases, both the real and imaginary parts of n_{eff} increase. This means that, toward the apex of the metal tip, the effective wavelength of the plasmon wave decreases and the losses increase. For radii of the metal tip larger than 50nm the losses are relatively low. As the radius decreases the losses become significant and they are minimum for wavelength around 800nm.

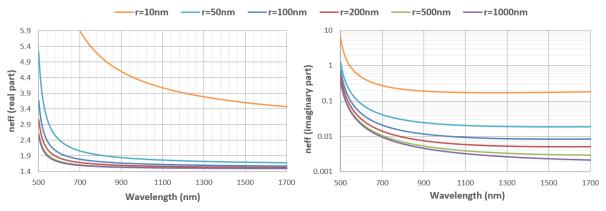


Figure 3. The real (left) and imaginary (right) part of the effective refractive index of TM_{01} mode for different radii of the gold core.

3.1 Simulations of metal tips

Simulations through Finite Element Method (FEM) using COMSOL Multiphysics were performed in order to study the enhancement at the apex of tapered hybrid microfibers. The simulated gold tapered core has input and output radius of 150nm and 3nm, respectively, and semi-angle of 3.5° . The cladding/core ratio is ~12 along the length of the fiber. Boundary mode analysis was used at the input of the fiber for the excitation of TM_{01} mode. Figure 4 (a) shows the enhancement spectrum of the tapered microfiber. The enhancement factor E_f was calculated as the maximum intensity at the apex of the tip divided by the intensity at the glass/metal interface at the input of the fiber. The maximum E_f calculated was ~2.5 x 10⁵ for 1100nm excitation wavelength. The normalized electric field for 1100nm excitation wavelength and its enhancement toward smaller radii is shown in Figure 2 (b). When the length of the tip is larger than 1µm the propagating SPPs are reflected at the tip's end and form cavity modes appeared as periodic peaks in the enhancement spectrum.

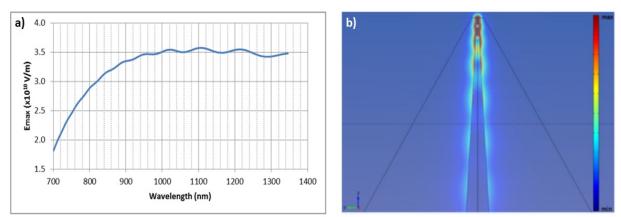


Figure 4. a) Enhancement spectrum of a tapered hybrid microfiber of $a=3.5^{\circ}$ and input and output radius of 150nm and 3nm, respectively. b) Normalized electric field for 1100nm excitation wavelength along a tapered microfiber.

4. EXPERIMENTAL

4.1. Tapering process

For the tapering of the microfibers the method of heating and stretching in a flame was used as shown^{8,9}. A schematic representation of the tapering setup is shown in Figure 5. The tapering rig consists of three stages, one that moves the micro-burner and two for stretching the fiber. The temperature profile and geometry of the micro-burner flame is tailored by mixing oxygen and butane gasses at appropriate flow rates. Two electronic mass flow controllers are employed for accurately controlling the oxygen and butane gas flow rates. The whole tapering process is computer controlled in order to achieve the desired final taper geometry.

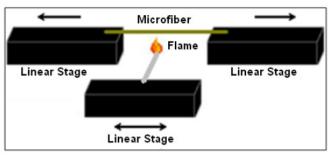


Figure 5. Schematic representation of the tapering rig.

The accurate temperature control is important in order to obtain a smooth hybrid tip. The appropriate tapering temperature is just below the melting point of gold (1064° C). For higher temperatures the gold core melts leading to the formation discontinuous core metal patterns or even the formation of well defined microspheres as can be seen Figure 6 (a). For the tapering process a whole range of different microfibers with continuous cores were used but the typical dimensions of the majority of fibers had metal core and silicate cladding diameters of ~10 μ m and ~35 μ m, respectively. For discontinuous cores and temperatures below the melting point of gold it was observed that the glass cladding collapses as shown in Figure 6 (b).

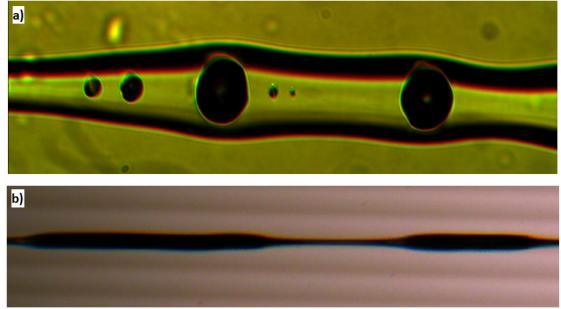


Figure 6. a) Hybrid microfiber with melted core due to tapering at $T > 1064^{\circ}$ C. b) Collapsed hybrid microfiber due to tapering at $T < 1064^{\circ}$ C.

For temperature just below 1064° C smooth tips were fabricated (Figure 7). The initial core/cladding diameter ratio is not preserved creating a sharp tip with a thick glass cladding facilitating the required robustness for easy handling.

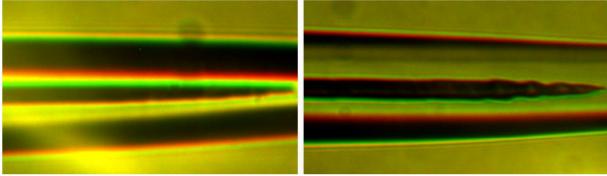


Figure 7. Fabricated tips.

In the following Figure 8 is demonstrated the drastic decrease of the metal core to silicate cladding diameter ratio. At the left picture is illustrated the initial untapered microwire where their ratio is about 1/3. At the right SEM picture it can be seen the tapered fiber tip where now the ration has been drastically decreased to 1/20.

In the tapered microwire's tip image the metal core could not be seen and also the core's shape looks asymmetric and non circular due to the non perfect cleaving that was attempted to the microwire.

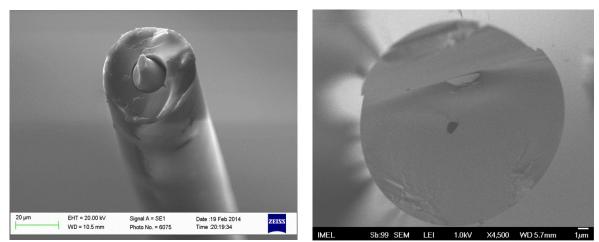


Figure 8. SEM picture of the initial microwire (left). SEM image of the tapered microwires tip which demonstrates the drastic decrease in core /silicate diameters ratio.

5. CONCLUSIONS

Fabrication of hybrid metal core tips by tapering metal core/dielectric cladding microfibers is proposed for high field enhancement and confinement. The hybrid microfibers used, offer ease of light coupling, as well as mechanical robustness. Even though a solitary spool up to 50 km in length of continuous diameter fiber can be fabricated, a few core discontinuities apparent in the entire length do not influence the TM_{01} mode profile due to the multiple re-excitation of the plasmon modes due to the repeatable glass/air total internal reflection. The simple and cost – effective method of heating and stretching in a flame was used for the tapering of the microfibers offering control of the tip's geometrical characteristics. For the tapering process though continuous cores with smooth surfaces are needed in order to get a good quality tip. Tapered tips with final core to silicate diameter ratio of 1/20 were demonstrated.

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REFERENCES

- [1] Stockman M. I., "Nanofocusing of optical energy in tapered plasmonic waveguides," Phys. Rev. Lett. 93(13), 137404 (2004).
- [2] Uebel P., Bauerschmidt S. T., Schmidt M. A. and Russell P. St J., "A gold-nanotip optical fiber for plasmonenhanced near-field detection," Appl. Phys. Lett. 103(2), 021101 (2013).
- [3] Zaccaria R. P., Alabastri A., De Angelis F., Das G., Liberale C., Toma A., Giugni A., Razzari L., Malerba M., Sun H. B. and Di Fabrizio E., "Fully analytical description of adiabatic compression in dissipative polaritonic structures," Phys. Rev. B 86(3), 035410 (2012).
- [4] Petropoulou A., Antonopoulos G., Bastock P., Craig C., Kakarantzas G., Hewak D.W., Zervas M.N. and Riziotis C., "Engineering of composite metallic microfibers towards development of plasmonic devices for sensing applications," IOP Conf. Series: Materials Science and Engineering 108, 012027 (2016).
- [5] Gholipour B., Nalla V., Bastock P., Khan K., Craig C., Hewak D. W., Zheludev N. I. and Soci C., "Plasmonic Nanowire Continuum Light Source," CLEO '14 (2014).

- [6] Bastock P., "Manufacturing Novel Fibre", Doctor of Philosophy, Optoelectronics Research Centre. University of Southampton (2015).
- [7] Handapangoda D., Premaratne M., Rukhlenko I. D. and Jagadish C., "Optimal design of composite nanowires for extended reach of surface plasmon-polaritons," Opt. Express 19(17), 16058 -16074 (2011).
- [8] Birks T. A., Wadsworth W. J. and Russell P. St. J., "Supercontinuum generation in tapered fibers," Opt. Lett. 25(19), 1415-1417 (2000).
- [9] Kakarantzas G., Leon-Saval S. G., Birks T. A. and Russell P.St.J., "Low-loss deposition of solgel-derived silica films on tapered fibers," Opt. Lett. 29(7), 694-696 (2004).