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Surface plasmon polariton excitation by second harmonic generation in single organic nanofibers

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Abstract: Coherent local excitation of surface plasmon polaritons (SPPs) by second-harmonic generation (SHG) in individual aligned crystalline organic functionalized para-phenylene nanofibers deposited on a thin silver film is demonstrated. The SH-SPP generation is considered theoretically and investigated experimentally with angular-resolved leakage radiation spectroscopy for normal incidence of the excitation beam. Both measurements and simulations show asymmetric excitation of left- and right-propagating SH-SPPs, which is explained as an effect of fiber molecules being oriented at an angle relative to the silver film surface.

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1. Introduction

Research efforts in the field of plasmonics and, particularly, in the development of plasmonic structures have increased rapidly over the past decade due to the potentials for fabrication of very compact circuitry. Development of active surface plasmon based components is required to sustain further advances in both fields. To integrate the plasmonic components into photonic components for datacom and optocom applications, development of active components that can convert optical signals into plasmonic signals and *vica versa* are needed. This demand stimulates a search for configurations, in which Surface Plasmon Polaritons (SPPs) can be locally excited in a well-defined part of the plasmonic circuitry. Several ways to achieve this have already been demonstrated [1,2], including both passive methods like ridge, slit, or grating couplers [3–5], and active methods based on e.g. four-wave mixing [6] or electrical excitation [7].

We have previously demonstrated that organic nanowires deposited on a thin silver film can act as local nonlinear sources that scatter second harmonic light into plasmons propagating along the silver film [8]. This method has the advantage that the generated SPPs are coherent with the excitation light, and that near-infrared optical signals are converted to SPP signals in the visible range, where they are easier to detect or convert afterwards. Also, the SPPs excited using this approach, have a well-defined wavelength that is tunable, the method does not require specific angles of incidence of the excitation light to work, and SHG in a single fiber offers an extremely local source of SPP generation compared to methods found in [1,2]. Furthermore, for near-infrared excitation, a considerable part of the generated light is coupled to SPPs, which leaves only a weak background signal at the SH frequency. However, this was demonstrated for an ensemble of wires. To take full advantage of the compact plasmonic structures that can be fabricated it will be an advantage to be able to use a single wire as the source for generation of plasmons. Here we demonstrate excitation of SPPs on an air/silver interface using second harmonic generation (SHG) in single 1-cyano-p-quaterphenylene (CNHP4) crystalline organic nanofibers deposited on the metal surface. It is shown that a nonlinear source region with a size of the order of the focal spot of the excitation laser ($\sim 2 \mu\text{m}^2$) can generate SPPs that are detectable in leakage radiation spectroscopy. While SPP generation from an ensemble of fibers showed symmetrical SPP generation in two directions along the surface the present experiments on single fibers show strongly asymmetrical SPP generation due to asymmetry in the orientation of the polar molecules in the fibers. The investigations thus reveal that generation of SPPs from a single fiber is very sensitive to the structure of the fiber which even for symmetric excitation conditions may lead to non-symmetrical generation of SPPs in two directions along the surface.

2. Theoretical study of leakage radiation from a single fiber

We have calculated both linear scattering and second harmonic generation (SHG) from a single CNHP4 fiber with a few different widths W and height 100 nm placed on a 40 nm silver film on a quartz substrate [Fig. 1] using the Greens function area integral equation method [8]. The geometry considered is similar to Refs [8,9]. but with slightly different fiber dimensions, and, while in [8] the fiber molecules were assumed to be parallel with the silver surface ($\alpha = 0^\circ$), here they are assumed to be at an oblique angle ($\alpha = 25^\circ$). The oblique angle is crucial for describing asymmetric SPP excitation in SHG observed also experimentally in later sections.

In linear scattering the fiber is illuminated from the air side with a normally incident plane wave of wavelength 390 nm. Figure 1(a) shows the power scattered into the quartz substrate

per unit angle as a function of angle θ for fibers of widths $W = 300$ nm and 350 nm. We observe a large central peak around $\theta = 0^\circ$, and two small peaks at app. $\theta = \pm 50^\circ$. The latter peaks are due to scattering of light into SPP waves that propagate along the air-silver interface, and then leak into phase-matched waves in the quartz substrate at angles given by [9,10]

$$k_0 n_{\text{quartz}} \sin \theta \approx \text{Re} \left(k_0 \sqrt{\frac{\epsilon_{Ag}}{1 + \epsilon_{Ag}}} \right). \quad (1)$$

As in [8] we have used the refractive index of quartz $n_{\text{quartz}} = 1.5$, the refractive index of CNHP4 fibers 1.65, and the dielectric constant of silver ϵ_{Ag} was obtained from [11]. The finite width of SPP peaks [Fig. 1] is related to the SPP propagation loss caused by both absorption in the silver film and leakage radiation into the substrate. The case of fiber width 400 nm gave almost vanishingly small SPP peaks (not shown). SPP leakage radiation peaks are still small for the slightly different fiber dimensions considered here [Fig. 1(a)] in agreement with experimental observations presented in the next section. Larger SPP peaks are obtained by choosing other fiber dimensions. Notice also the symmetry of linear scattering spectra, which is a consequence of using normally incident light.

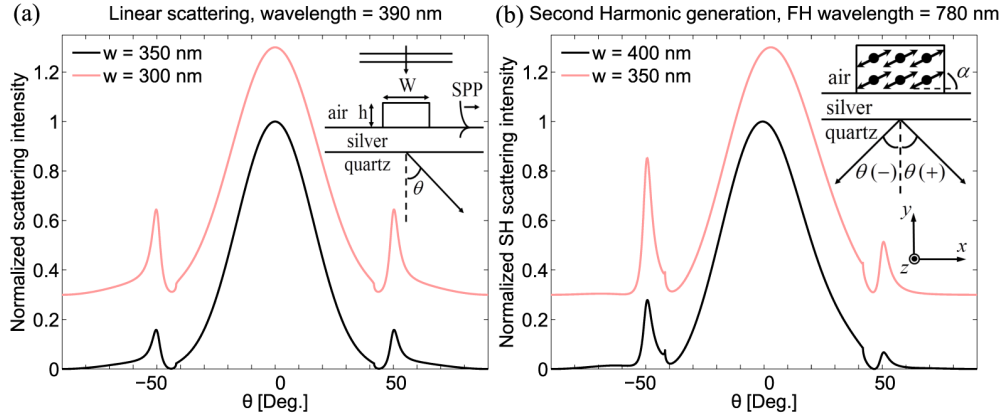


Fig. 1. Calculated angular leakage radiation spectra for a CNHP4 fiber of width W and height 100 nm placed on a 40 -nm-silver-film-on-quartz geometry, and being illuminated with normally incident light from the air side with electric field polarized along the x axis. (a) Linear scattering, and (b) Second Harmonic generated power, per unit angle versus angle in the quartz substrate. For linear scattering the pump wavelength is 390 nm, and for SHG it is 780 nm but the detected SHG power is at the wavelength 390 nm. The molecules of CNHP4 fibers are oriented at an angle of 25 deg. relative to the silver surface. Red curves are shifted by 0.3 on the second axis for clarity.

For the case of SHG [Fig. 1(b)] the fiber is illuminated with a normally incident plane wave of wavelength 780 nm. A complete calculation of linear scattering at this wavelength was used to find the fundamental harmonic (FH) electric field inside the CNHP4 fiber. We then assumed the fiber molecules to be at an angle of $\alpha = 25$ deg. relative to the silver surface [see inset, Fig. 1(b)], and that SHG is generated by a Second Harmonic polarization given by:

$$\mathbf{P}(\mathbf{r}, \omega_{SH}) = \chi^{(2)} (\hat{x} \cos \alpha + \hat{y} \sin \alpha) P(\mathbf{r}), \quad P(\mathbf{r}) = [(\hat{x} \cos \alpha + \hat{y} \sin \alpha) \cdot \mathbf{E}(\mathbf{r}, \omega_{FH})]^2, \quad (2)$$

where $\chi^{(2)}$ governs the strength of the SHG process and is dominated by the molecular dipoles as shown in [12]. This constant does not affect the shape of angular spectra and can be set to 1 for our purpose for positions inside the fiber, and 0 otherwise. Note that previous work for an

ensemble of fibers only considered the case $\alpha = 0$ [8]. We have then used the expression (2) as a given polarization at the SH wavelength, and again solved the linear scattering problem at the SH wavelength taking into account that radiation generated by the given SH polarization will propagate to other places inside the fiber, possibly mediated by reflection from the surface, which will lead to fields there that induce an additional linear polarization at ω_{SH} .

The additional calculation step is carried out by solving

$$\mathbf{E}(\mathbf{r}; \omega_{SH}) = \int \mathbf{G}(\mathbf{r}, \mathbf{r}'; \omega_{SH}) \frac{\omega_{SH}^2}{c^2} \cdot \left[\frac{\mathbf{P}(\mathbf{r}'; \omega_{SH})}{\epsilon_0} + (\epsilon(\mathbf{r}'; \omega_{SH}) - \epsilon_{ref}(\mathbf{r}'; \omega_{SH})) \mathbf{E}(\mathbf{r}'; \omega_{SH}) \right] dx' dy', \quad (3)$$

which also emphasizes that the electric field at the SH frequency $\mathbf{E}(\mathbf{r}; \omega_{SH})$ is generated by both a given polarization and an additional induced polarization. \mathbf{G} is the Greens tensor for the layered reference geometry, ϵ is the dielectric constant, and ϵ_{ref} is the dielectric constant of the layered reference geometry (see [8] for details). The asymmetric SHG spectrum for angles $|\theta| > 41.8$ deg. (critical angle of air-quartz interface) shown in Fig. 1(b) is not a trivial result, and asymmetry does not automatically follow above the critical angle when $\alpha \neq 0$. For a fiber with a very small cross section we find two SPP peaks with identical heights no matter the choice of α (not shown). This can be explained similar to Ref [9], where it was theoretically shown for small fiber cross sections that the angular spectra will be symmetric above the critical angle of the air-quartz interface for a total polarization (given and induced) on the form of Eq. (2) no matter the value of α . Note that Eq. (2) describes linear polarization. For elliptical polarization, asymmetric SPP excitation is on the other hand possible [9,13]. Symmetric angular spectra will also be the case for fibers of large cross section if we ignore the second term in the square parenthesis of Eq. (3) and assume that $P(\mathbf{r})$ is mirror symmetric across the plane with fixed x at the center of the fiber. This follows from properties of the far-field expression of \mathbf{G} [9]. Mirror symmetry of $P(\mathbf{r})$ follows for normal incidence if the resulting $\mathbf{E}(\mathbf{r}; \omega_{FH})$ is predominantly oriented along the x -axis. However, if there is a y -component of $\mathbf{E}(\mathbf{r}; \omega_{FH})$ then it can be shown that it will have opposite sign in opposite ends of the fiber and thereby cause an asymmetry in $P(\mathbf{r})$ for non-zero α according to Eq. (2). Note also that if Eq. (2) is modified to include partial molecule orientation along the z -axis this leads to an additional component in angular spectra with no SPP excitation from s -polarized SH fields (not shown).

3. Materials and experimental methods

The sample used in the experiment consists of a 0.5 mm thick glass substrate (10 x 10 mm square) coated with a 40 nm silver film (with a roughness of approximately 1.5 nm) on one side. The CNHP4 fibers were grown on mica and soft-transferred to the air/silver surface of the sample [14]. To reduce the bleaching of the SH signal due to photo-oxidation, a SiO_x layer was added onto the fiber side of the sample [15]. The average dimensions of the fibers were measured with atomic force microscopy to be approximately 100 nm high, 400 nm wide and 20 μm long with an average spacing of 5 μm . To excite the SPPs a femtosecond laser system (Spectra-Physics Tsunami Ti:Sapphire oscillator) was used to deliver ~ 85 fs pulses at a repetition rate of 80 MHz at 785 nm. The power and polarization of the light reaching the sample was controlled by neutral density filters and a pair of polarizers. In the experiments light polarized in the scanning plane of the detector was used (p -polarization). As illustrated in Fig. 2a the sample was mounted onto a cylindrical glass prism with an index matching liquid in between, so the long axes of the fibers were perpendicular to the scanning plane of the detector. In this configuration, where p -polarized light was used, the fibers were generating maximum SHG light [16]. The back reflection from the sample was focused onto a CCD camera, giving an image of the area investigated. To ensure that the laser focus was on a

single fiber, and to check for any defects on the fibers, a SH image was recorded by raster scanning the 2D stage mounted sample holder, and the SH light generated from the free propagating modes transmitted through the silver film in the forward direction was detected (the detector positioned at $\theta = 0^\circ$, Fig. 2(a), with almost no two photon luminescence for the wavelength used [12]). The light was focused onto the sample by a microscope objective (40.0X, N.A. 0.65) giving a spot size of $1.5\mu\text{m}$ (full width half maximum) and an average power density of approximately 20 W/cm^2 , which was below the damage threshold of both the fibers and the silver film. A colored glass filter (RG715) was placed before the objective to remove any SH light generated in the optics before the objective, and after the sample a BG39 bandpass filter was placed in order to remove any residual excitation light as well as any third-harmonic light generated. To reduce the angle range from which light was collected to 2 degrees, an adjustable slit was placed between the sample and the photomultiplier tube (PMT) used as detector.

4. Linear scattering by a single nanofiber

A frequency-doubled light beam of wavelength 392.5 nm (785 nm frequency doubled in a BBO crystal) was focused onto a single fiber acting as a scatterer near the silver surface. The angular distribution of light exiting the quartz prism shows a broad symmetric feature at small angles and two features at large angles (red curve in Fig. 2(d)). The observed symmetry of the red curve could not generally have been expected if the linear material response had been anisotropic, and thus the observed symmetry supports that we can use a scalar dielectric constant in Eq. (3). The missing part at the center of the broad central feature is due to the beam block protecting the detector from direct exposure to the transmitted part of the excitation beam. Figure 2(d) also shows leakage radiation spectra for linear scattering of the frequency-doubled beam for an ensemble of fibers, and SHG from the same single fiber with the excitation source of 785 nm discussed in next section. As expected, two symmetric peaks appear in the spectrum in the case of the linear scattering process, since this is independent of the orientation of the molecular dipole moment.

Comparing the spectra from scattering on a single fiber with the numerical analysis found in Fig. 1(a), the experimental plot indicates more weak features. The explanation is found in scattering due to the surface roughness of the air/silver interface (not included in the linear or SH simulations) causing a broadening of the SPP peaks, i.e. weaker and broader features are observed.

It is noticed that the position of the SPP peak is slightly shifted towards lower angles in the experiments compared to simulations. This is caused by the finite thickness of the 0.5 mm glass substrate causing a vertical displacement of the silver film and fibers compared to the center of curvature of the cylindrical prism, which leads to refraction of leakage-radiation light when reaching the prism-air surface (also observed in the SH measurements).

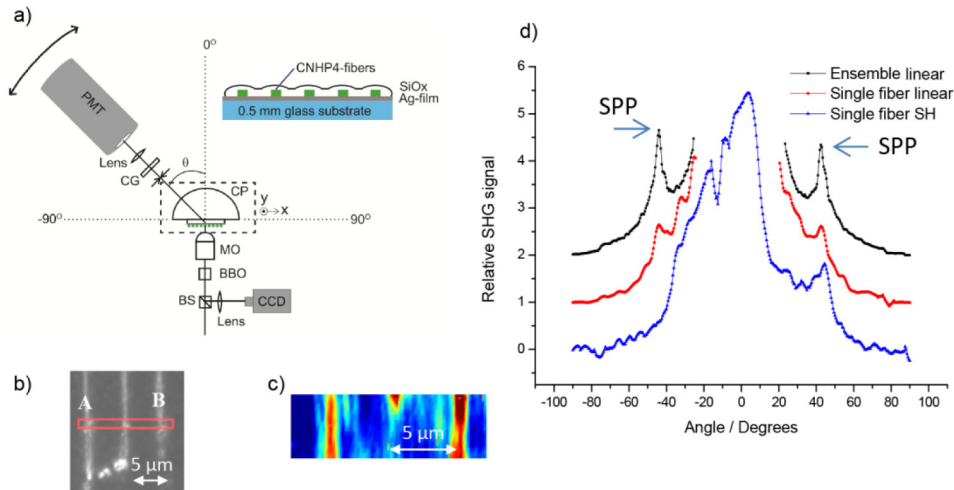


Fig. 2. a) Schematic of the experimental setup used: BS: Beam splitter, MO: Microscope objective, CP: Cylindrical prism, CG: Colored glass filter, SiO_x: 50 nm SiO_x layer, Ag-film: 40 nm silver layer, PMT: Photo Multiplier Tube, CCD: Charge Coupled Device camera, BBO: Beta Barium Borate crystal used in the linear scattering experiments. The insert shows: A schematic of the sample mounted on the flat surface side of the cylindrical prism (up-side down). b) Microscope image of the investigated fibers. The red box illustrates the area rasterscanned. c) SH image of the rasterscanned area. One should note the defect in the fiber between A and B. d) Three different angle resolved leakage radiation spectra. Linear scattering with an excitation source of 392.5 nm (785nm frequency doubled in a BBO) on a single fiber (red curve), SH light generated for the same single fiber (blue curve), and the linear scattering on an ensemble of fibers (black curve). All curves are normalized separately.

5. Excitation of SPP by SH light generated in single fiber

FH light was focused onto one single fiber at a time (fibers A or B, Fig. 2(b)), and SH light generated inside the fiber due to the non-zero second order susceptibility of CNHP4 [16] was used to excite SPPs. The angular distribution of light exiting the quartz prism was measured for 785 nm excitation wavelength. Figure 3(a) shows the relative SHG signal as a function of the scanning angle, together with a linear scattering for an ensemble of fibers for clear identification of the SPP peak position. Two features dominate the spectrum of the fibers A and B: A broad central peak produced by SH light generated in the form of free propagating modes that are partially transmitted through the silver film in the forward direction, along with a peak corresponding to leakage radiation from generated SPPs located at an angle higher than the critical angle of an air-glass interface [10]. Comparing the angle resolved leakage radiation scans for the two fibers A and B, one can see that for the fiber A the SH-SPP peaks are suppressed for negative angles whereas for the B fiber it is for positive angles. This suggests that A and B have different molecular growth directions. Such asymmetry was also observed in the simulations in the previous section [Fig. 1(b)]. Experiments on CNHP4 fibers with no SiO_x layer shows that the angle between the substrate and the molecule orientation of the fiber was $7 \pm 5^\circ$ [16], but adding a layer of SiO_x coating on the organic fibers shows that the molecules become more likely to bend away from the surface because of an increased stiffness, and therefore 15-20° angles was expected, as observed with SEM for other organic fibers [15]. Earlier experiment found in [8] on an ensemble of CNHP4 fibers for samples with no SiO_x shows two almost symmetric SH-SPP peaks. This is simply because the average over a large number of different fibers is measured [17].

Figure 2(d) compares the leakage radiation spectra for linear scattering of the frequency-doubled beam with the SHG from the same single fiber with the excitation source of 785 nm. One can see that the asymmetric SPP excitation only appears when a single nanofiber acts as

the source for generating the SH-SPPs. This confirms that the SH-SPP peaks originate from the coupling of SH light generated inside the CNHP4 nanofibers. Contrary to the SH-SPP peaks the asymmetry in the central peak in Fig. 2(d) may be caused by asymmetry in the cross section of the investigated fiber.

In order to check the influence of the laser focus position on the fiber and the SH contribution from the Ag film, a measurement was performed where the laser focus was moved perpendicular to the long axis of a fiber. Here the asymmetric behavior does not change, and further scans with the laser focus positioned at $\pm 0.4 \mu\text{m}$ and $\pm 0.5 \mu\text{m}$ show the same tendency with a lower signal to noise ratio, and for measurements on bare Ag-film the SPP peak is not seen in the spectrum. Therefore the asymmetry is due to the orientation of the dipole and the complex morphology of the nanofiber. Comparing the red curve in Fig. 3(b) with Fig. 1(b) one should note that the SPP peak looks more symmetric in Fig. 3(b). Since the fibers on the sample are not identical, the dimensions and the angle α can differ, resulting in a different spectrum for different fibers. In this particular case α looks smaller than 25° which was used in the simulations.

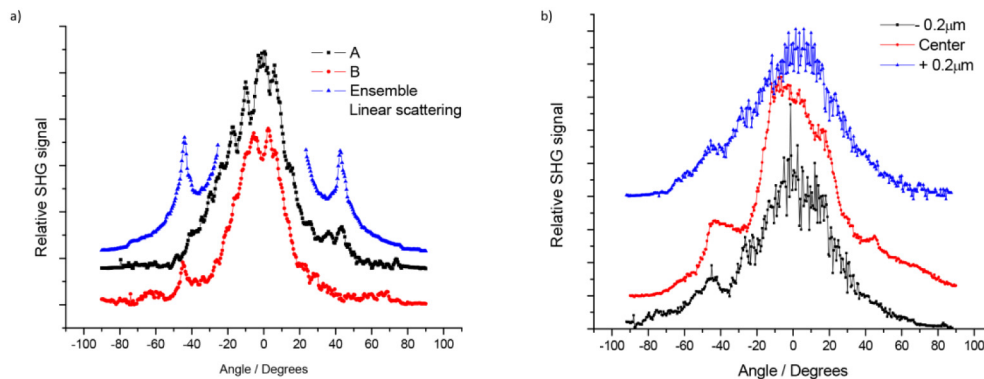


Fig. 3. a) Angle resolved leakage radiation spectra with the excitation source of 785 nm, where curves A and B represent the SPP excitation by SH light generated in two different fibers in Fig. 2(b). The blue curve: Linear scattering with an excitation source of 392.5 nm (785 nm frequency doubled in a BBO crystal) on an ensemble of fibers. All curves are normalized separately. b) Focus scan of a SHG signal normalized after the SPP peak, the red curve was measured when the laser focus was centered on a single fiber in the direction perpendicular to the fiber, whereas the blue and black were measured for respectively $+0.2 \mu\text{m}$ and $-0.2 \mu\text{m}$ displacement. Note that the signal to noise ratio decrease in the two non-centered measurements is due to the reduced overlap between the laser beam and the fiber.

6. Conclusions

The angle resolved leakage radiation spectra in Figs. 2(d) and 3(a) clearly demonstrate SPP excitation achieved through SHG in single nanofibers deposited on a smooth silver surface. In the measurements an asymmetric behavior of the SH-SPP coupling was observed for single fibers. Theoretical simulations showed that the asymmetry can be related to the angle of the molecular dipole moment of the fiber molecules relative to the silver surface. We have shown that the fibers supply the optical non-linearity necessary for efficient SHG, and SH-SPP excitation will be possible in very close vicinity of the fibers, which therefore can be used as an extremely local SPP source. Combined with earlier studies, this work further supports that the SH-SPP excitation is a promising alternative for creating tunable coherent local SPP sources, e.g. for surface plasmon circuitry.

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