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Excitation of fluorescent nanoparticles by channel plasmon polaritons propagating in V-grooves

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Recently, it has been proven that light can be squeezed into metallic channels with subwavelength lateral dimensions. Here, we present the study of the propagation of channel plasmon polaritons confined in gold V-grooves, filled with fluorescent particles. In this way, channel plasmon polaritons propagating in nonempty V-grooves can be characterized, as the propagation track can be directly visualized in the microscope. We have found that beads with subwavelength diameters act as frequency converters for the propagating channel modes, resulting in larger propagation lengths. For micrometric-diameter beads, we show the possibility of individual excitation, what may have applications to develop very sensitive biosensors. © *2009 American Institute of Physics*. [doi:10.1063/1.3262945]

Surface plasmon polaritons (SPPs) are evanescent waves resulting from the interaction between an electromagnetic field and the electrons in the conductive band of a metal.[1,2] They are bounded to and propagate along the metal/dielectric interface. The properties arising from their hybrid nature (e.g., shorter wavelength) can be exploited to overcome the diffraction limit and to squeeze light in submicrometric optoelectronic devices[3,4]

SPPs that are laterally confined in subwavelength structures (e.g., in metal slits or gaps) are called channel plasmon polaritons (CPPs).[5–7] As a consequence of the confinement, CPPs present a shorter propagation length than SPPs excited in similar conditions.[6] However, the simultaneous confluence of strong confinement and a propagation loss sufficiently low for practical applications has been long out of reach. Recently, a promising option for the realization of effective plasmonic waveguides based on V-shaped metallic grooves has been proposed.[8–12] In comparison with other configurations, they show superior features, such as strong localization of the CPP modes, relatively low propagation losses even through sharp bends,[9,11] low sensitivity to surface roughness or defects, and broadband transmission.[11]

A theoretical study of such structures was reported by Pile and Gramotnev,[8] and pioneer experimental measurements were performed by Bozhevolnyi *et al.*[10,11] Characterizations by scanning near field optical microscopy (SNOM) in V-grooves fabricated by direct milling of the metal demonstrated subwavelength CPPs confinement. The propagation lengths were 90–120 μm at wavelengths of $\lambda_0 \approx 1.5$ μm. Recently, Vernon *et al.*[13] calculated theoretically the effects of filling the V-grooves with a dielectric ($\varepsilon_d > \varepsilon_{air} = 1$). The main consequence is an increase in the localization of the CPP, leading to a shorter propagation length and to an increase in the minimum dimensions of the grooves necessary to support CPPs. Thus, V-grooves filled with a dielectric can have an impact on their application as waveguides: a higher confinement allows bending light with lower losses, and the geometrical conditions for CPP existence are less strict, facilitating the design and fabrication of devices. However, no experimental results have been reported up-to-date, as SNOM characterization is not possible in nonempty grooves. In this work, we report on experimental investigations of CPPs propagating in a V-groove filled with a fluorescent dielectric material. This allows studying the propagation mechanism and parameters by using a conventional optical microscope.

Devices consisting on several V-grooves (200 nm thick gold layer on a transparent polymer substrate) with different lengths (from 100 to 500 μm) were used for this purpose. Recently, we have developed a method for the massive fabrication of such devices, based on nanoimprint lithography.[14,15] The process allows to integrate in the same device several V-grooves with deep channels to perform optofluidic and fiber-to-fiber experiments. Grooves with various widths (2–8 μm) and a fixed apex angle of $\theta \approx 50°$ were fabricated and optically characterized using SNOM. Figure 1(a) shows a schematic of the device and a scanning electron microscopy (SEM) image of a V-groove. SNOM characterizations were reported,[15] demonstrating subwavelength confinement and guiding with relatively low propagation losses. The measured propagation lengths at $\lambda_0 \approx 1.5$ μm were ~90–140 μm, which are similar to the values observed in focused ion beam-milled devices.[10,11,16]

Nanometric size, fluorescent polystyrene beads were deposited by capillarity, completely filling the V-grooves, as can be seen in the SEM image of Fig. 1(b). Fluorescence images (overall illumination) revealed a uniform distribution along the grooves. The beads were 100 nm diameter (ϕ), red fluorescent, with an excitation maxima at 542 nm (green),

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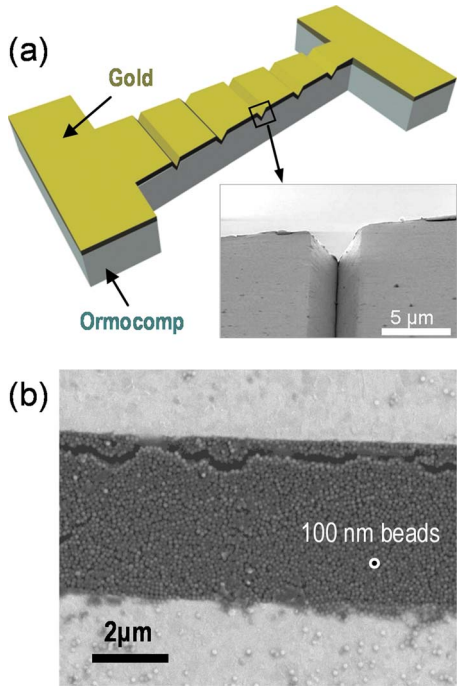

FIG. 1. (Color online) (a) Sketch and SEM image of a device based on several V-groove waveguides, made in gold (200 nm) onto a transparent polymeric substrate (Ormocomp) by a nanoimprint-based process. (b) Image of a detail of a V-groove filled with 100 nm diameter red fluorescent beads.

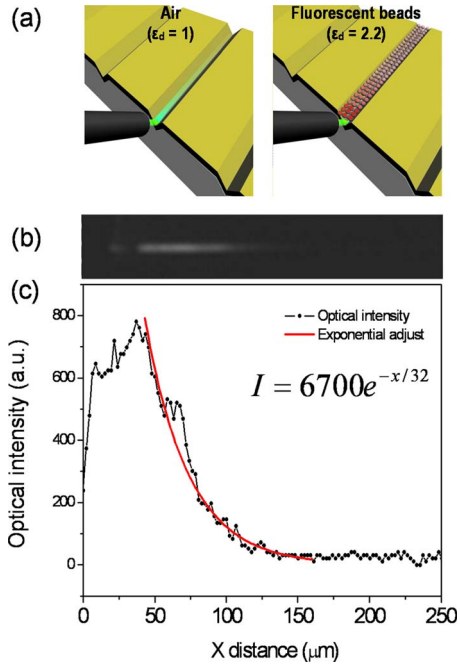

FIG. 2. (Color online) (a) Sketch of the experimental set-up: the incident light is launched at one end of the V-groove, with the sample tilted with respect to the fiber to avoid direct excitation. The beads convert the near field of the excited CPPs to far field, by the emitted fluorescence. The beads form a compact structure, so the filling material can be described by $\varepsilon_d = 2.2$, thus, allowing studying propagation in nonempty grooves (i.e., $\varepsilon > 1$). (b) Top view far field image, obtained with an optical microscope, of the light emitted by the fluorescent nanoparticles excited by the CPPs propagating in a filled V-groove. (c) Optical intensity profile obtained by averaging the signal of (b) and its exponential fit, from where a propagation length of 32 μm is estimated.

and an emission maxima at 612 nm (red). The nanoparticles occupy 80% of the available volume (squared-volume filled with self-assembled spheres), so the media can be approximated as a weighted average of air (0.2) and polystyrene (0.8). Thus, an effective refractive index can be estimated: $0.2 \cdot n_{air} + 0.8 \cdot n_{polystyrene} = 1.4$ (i.e., the effective dielectric constant is $\varepsilon_d = 2.2$). For the wavelengths of the laser source used in this work, e.g., $\lambda_0 = 532$ nm (close to the excitation maxima), the corresponding wavelength in medium is $\lambda = \lambda_0 / 1.4 = 380$ nm. Hence, the beads diameter is smaller than $\lambda/2$ for the shortest wavelength used so that the medium inside the grooves can be approximated as a continuous dielectric.

After filling the V-grooves, the excitation of CPPs was done by illuminating locally one end of the V-grooves, with the fiber tilted with respect to the sample (to minimize the direct lightening of the beads) as it is shown in the drawings of Fig. 2(a). The *top view* images were obtained with a camera coupled to a conventional optical microscope using adequate filters, so only the red (fluorescent) light was recorded. Figure 2(b) shows the fluorescence image of a V-groove illuminated with a green laser ($\lambda_0 = 532$ nm) on the left side. The graph in Fig. 2(c) corresponds to the optical intensity profile along the V-groove averaged from (b) and its exponential fit. A propagation length of $\delta_{CPP} \approx 32$ μm was estimated from the exponential fit.

The dependence on the polarization was studied as well. For this, a broadband source in the visible range was used to excite the beads. A polarizer was placed in-between the light source and a polarization-maintaining fiber, used to launch the polarized light into the V-grooves. The maximum confinement was observed to occur for TE polarization (the electric field parallel to the flat surface), as this satisfies the condition for CPPs excitation. Unexpectedly, a propagation length of $\delta_{CPP} \approx 33$ μm was also measured for this white source.

A significant decrease of δ_{SPP} when increasing ε_d was expected and already predicted in[13] as a consequence of the stronger confinement. But there are two experimental observations that point to a more complex mechanism of CPP propagation in the present system.

(I) The first one is that the δ_{CPP} observed for the CPPs excited with the green laser is longer than the calculated for SPPs excited in similar conditions (green laser, gold/beads interface) using Eq. (1),[17] that is $\delta_{SPP} = 23$ μm (considering $\varepsilon_d = 2.2$, as described above, and $\varepsilon_m = -14.5 + 0.18i$, according to Drude´s model for $\lambda_0 = 532$ nm). Theoretical ε may be very different from that of real materials, especially in metals, but, in any case, the propagation length in a real system should be shorter than the theoretical one, as the propagation losses are higher due to defects or dust, poor quality of the structures and/or the materials,

$$\delta_{SPP} = \lambda_0 \frac{(\varepsilon_m')^2}{2\pi\varepsilon_m''} \left(\frac{\varepsilon_m' + \varepsilon_d}{\varepsilon_m' \varepsilon_d} \right)^{3/2}. \quad (1)$$

(II) The second one is that the propagation length is the same for the visible broadband source and for the green laser, which is counterintuitive, as the propagation length strongly depends on the incident λ_0.

According to these observations, we infer that the fluorescent nanometric beads are acting as frequency converters for the CPPs propagating along the V-grooves. The light reemitted by the beads has a wavelength of 612 nm, which is

far above the excitation maximum needed to directly excite other beads, but that can create new CPPs in the V-grooves. These CPPs have an associated wavelength shorter than λ_0 as a consequence of the hybrid nature of these quasiparticles, so they can be absorbed by the beads. It does not exist a theoretical evaluation of the wavelength of CPPs propagating in a V-groove, but for SPPs excited at the conditions of our experiments (red light, gold/beads interface, i.e., $\varepsilon_m = -24.3 + 0.38i$ and $\varepsilon_d = 2.2$) the theoretical wavelength is $\lambda_{SPP} = 417$ nm, as calculated by

$$\lambda_{SPP} = \lambda_0 \sqrt{\frac{\varepsilon_d + \varepsilon_m'}{\varepsilon_d \varepsilon_m'}}, \tag{2}$$

which is below the excitation maxima. SPPs with this wavelength have an associated propagation length of $\delta_{SPP} = 38$ μm [calculated with Eq. (1)]. Then, the wavelength associated to the CPPs will be $417 \text{ nm} < \lambda_{SPP} < 612$ nm, indicating that it can be absorbed by the beads, as the absorption of the fluorescent beads can be approximated to be in the range of 542 ± 50 nm.

Thus, in both cases, there are two excitation sources: (1), light coming from the external source, from which only the green wavelengths can directly excite the beads. And (2), the CPPs excited by the light re-emitted by the beads, which associated wavelengths lie in the absorption range (green). Red light coming from the external broadband source may also excite CPPs which wavelengths can be absorbed by the beads, contributing to this effect. The propagation length associated to green excitation is shorter than that associated to red excitation. Thus, for both cases, the observed propagation length for CPPs in the V-grooves is associated with the excitation by red light. Hakala *et al.*[18] have recently reported the conversion ratio of SPPs by organic molecules in a silver surface to be high (~50%), which is in good concordance with our observations.

Micrometer size beads (1–3 μm diameter) were also deposited inside the grooves. When deposited in low concentrations, they could be observed individually [inset in Fig. 3(a)]. Figure 3 shows an image with ambient illumination (a), and the corresponding fluorescence nonfiltered images (b) and (c), showing the possibility of individual excitation. The beads were ~95 μm far from the incident spot which is in the limit of the propagation length. At these conditions, just by tuning the input optical intensity, the number of excited beads could be controlled, as it can be seen by comparing (b) and (c) in Fig. 3.

The ability of local excitation of CPPs and of light propagation inside sub-μ channels opens several possibilities that can be exploited for biosensing. Surface plasmon resonance (SPR) is a well known technique, which allows measuring changes in the refractive index at the vicinity of a metal surface. The geometry of the present devices (integrated V-grooves with deep channels) allows easily performing fiber-to-fiber measurements, thus, exploiting the high sensitivity of SPR, which would be probably higher when studying coupled modes. Sensitivity to external agents can be tuned by properly selecting materials to fill in the V-grooves. Also, individual excitation of fluorescent nanoparticles can be studied for the development of point-sources, or exploded by proper functionalization of the particle for single-protein detection.

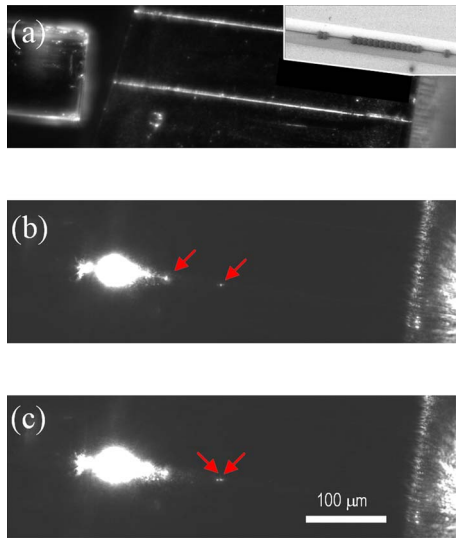

FIG. 3. (Color online) (a) Topography of a sample, where the fiber (at the left) and the v-shaped waveguides (horizontal bright lines) can be seen. Micrometric red fluorescent beads were deposited inside the V-grooves in very low concentrations, so they could be individually observed (SEM image in the inset). (b) and (c) correspond to fluorescence images (nonfiltered), were the individual excitation of beads is shown. The number of excited beads can be selected by tunning the intensity of the incident light.

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