Tunable nanowires: An additional degree of freedom in plasmonics

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We report on the excitation of surface plasmon polaritons (SPP's) on a periodic arrangement of metallic ribbons with tunable periodicity. The ability to vary its lattice constant results in an additional degree of freedom, permitting excitation of SPP's for any combination of wavelength and angle of incidence within the tuning range of the system. Moreover, the designed smart material allows fundamental questions to be answered regarding SPP propagation and localization. Finally, the concept opens vistas in the development of other methodologies in spectroscopy including plasmonic sensors and chips.

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I. INTRODUCTION

Planar waveguides and photonic crystal structures are being intensively investigated as key components for future integrated photonic devices. However, there may be an alternative approach to the manufacturing of highly integrated optical devices with structural elements smaller than the wavelength, which, nevertheless, enables strong guidance and manipulation of light—the use of metallic and metallodielectric nanostructures in conjunction with surface plasmon polaritons (SPP's). They are now considered as possibly "the next big step" in nanotechnology. Here, we present a device that allows freedom to choose the properties of excited SPP's to a certain degree at will. It is based on a smart, adaptive material which consists of a periodic arrangement of metallic, ribbonlike wires on a tunable polymer substrate, which allows tuning the periodicity of this structure over a wide range.

SPP's are mixed states of photons and electron density waves which propagate along the surface of a conductor. Their favorable properties allow strong guidance and manipulation of light on very small scales. This has led to a new branch of photonics called plasmonics.[1] Today SPP's are already playing an important role in the fundamental understanding of quantum behavior at nano- and mesoscales,[2–6] in subwavelength optics,[7] as well as in the development of ultra-surface-sensitive techniques like surface plasmon resonance,[8,9] and surface enhanced Raman scattering.[10]

Conventionally, SPP's are excited using an attenuated total reflection setup[11,12] or modulated metal surfaces with a fixed geometry.[11,13–17] Considering the latter case, the SPP excitation condition $k_{xl}^{light}(\omega)=k_x^{SPP}(\omega)$ can be approximated for a metal/air interface with a periodic modulation (periodicity d) by

$$\frac{\omega}{c}\sin(\alpha_{inc})+l\frac{2\pi}{d}\overset{!}{=}\frac{\omega}{c}\sqrt{\frac{\epsilon_m}{\epsilon_m+1}}, \tag{1}$$

where ϵ_m is the real part of the metallic dielectric constant, c is the vacuum velocity of light, α_{inc} is the polar angle of incidence, ω is the angular frequency, and l is an integer characterizing the diffraction order of the light in the periodic structure. Henceforth, an SPP which is excited using the lth diffraction order will be referred to as lth order SPP. The corresponding angle $\alpha_{inc}=\alpha_{SPP}$ will be called the resonance angle (of the lth order). Unfortunately, setups with fixed geometry confine and limit the experiment to a unique combination of frequency and incidence angle of the exciting light [see Eq. (1)]. In addition, for any given excitation frequency and angle, the properties of the SPP (as, e.g., its lifetime) are determined unambiguously.

These severe limitations are overcome by tunable gratings demonstrated in the present work. Tunable gratings not only allow selection of the incidence angle at will (e.g., normal incidence) but (as will be shown below) also give the freedom to tailor SPP properties such as group velocity and lifetime. Thus they give the possibility to isolate and study, e.g., ultraslow plasmons or to produce ultradispersive gratings[18] for a desired wavelength range. The tunable gratings consist of metallic wires separated by slits with adjustable width. Therefore, they can further be used to address fundamental questions connected with the transition of a propagating SPP excitation to a set of interacting but localized particle-plasmon excitations. Additionally, the geometry dependence of the plasmon-light coupling strength, (i.e., details of the SPP formation) can be studied systematically. In this paper, we want to present the proof of principle of such a device and show its impact on fundamental physics by comparing measurements and numerical simulations of SPP's on such systems.

II. TUNABLE SMART MATERIALS

The synthesis of the desired tunable structures is a multistep procedure described in great detail in Ref. 19. In brief, polydimethyl siloxane (PDMS) is cast to an approximately 1 mm thick rubber film. A uniaxial strain is then applied to the sample while an argon plasma truncates and cross-links the upper surface of the film. This leads, due to the fragmentation of macromolecular segments and renewed cross-linking, to relaxation processes in this thin surface region. After the strain is removed, the lower nonrelaxed part of the sample shrinks and causes compressive stress to the upper

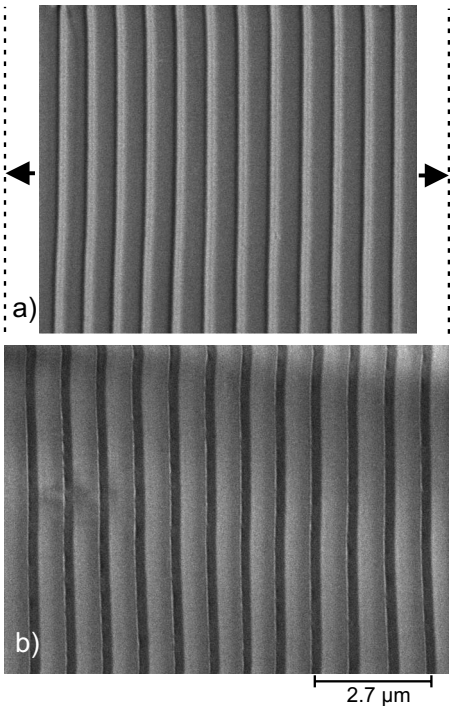

FIG. 1. Scanning electron micrograph of a silver coated periodic polymer structure with average silver thickness of 50 nm: (a) unstretched, periodicity 750 nm; (b) 20% stretched, periodicity 900 nm.

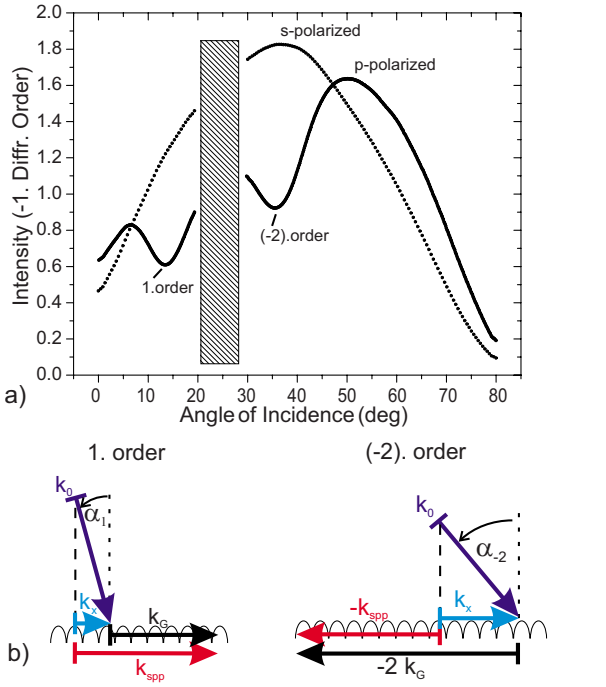

FIG. 2. (Color online) (a) Scan of the intensity of the −1 diffraction order versus angle of incidence of *s*-polarized and *p*-polarized light. Lattice constant is fixed at 780 nm. The experimental setup does not allow measurements in the shaded area. (b) **k**-vector diagram of the SPP excitation: 1st order (left) and −2nd order (right).

relaxed part. This produces sinusoidal ripples at the surface in order to compensate for the consequent length differences between the two layers. The sinusoidal profile is subsequently silver coated at an oblique angle of typically 45° in the plane perpendicular to the grooves, which results in a thickness modulated silver film at the surface. A scanning electron micrograph of such a surface is shown in (Fig. 1(a)).

Reapplication of strain leads to fractures along the thin metal regions parallel to the grooves. Such a film stretched by 20% is shown in Fig. 1(b). After an initial strain, a threshold is surpassed and, consequently, all the grooves break and the resulting metallic ribbonlike structures are almost perfectly aligned parallel to each other. The spacing, and hence fundamental periodicity of the grating, depends linearly on the applied strain. This was checked locally in the laser spot, as will be explained below. The separation of the metallic wires can be reversibly adjusted, thus allowing for dynamical changes in the grating constant. The initial value of the grating depends on the chosen PDMS, the stretching prior to the argon plasma treatment, and the plasma parameters. This procedure so far allows periodicities between 350 nm and 2 μm, while the separation can be fine tuned from 0 to above 50% of the initial periodicity.

III. TUNABLE SURFACE PLASMON EXCITATION

The silver/PDMS wire grating is used to demonstrate the tunability of the SPP resonance. As a first proof of principle, angle-dependent reflectivities for *s* and *p* polarizations have been recorded as a function of incidence angle α_{inc}. The light source used was a He-Ne laser (λ_0=632.8 nm). Figure 2(a) shows the reflectivities detected in the −1st diffraction order of an unstretched grating: the *s*-polarized curve does not show any minima, while the *p*-polarized curve has two pronounced minima at 15° and 36°. Note that only *p*-polarized light, with its polarization direction perpendicular to the wires, can excite SPP's. Therefore, we can conclude that these minima are caused by SPP's which extract energy from the propagating modes (light). The minima are found at the resonance angles, predicted by Eq. (1) for the 1st and −2nd order (l=1,l=−2) (ϵ_M=−15.87+1.08i). Having demonstrated SPP excitation, the next important step is to prove the promised tunability. Figure 3(a) shows reflectivity measurements for six different grating periods ranging from the unstretched sample to about 7% prolongation. The grating periods have been determined independently by using diffraction techniques (locally in the laser spot) and have additionally been verified by scanning electron micrographs. The two minima of the 1st and −2nd order plasmons are clearly visible in each case. Their positions coincide reasonably well with the predictions of Eq. (1). Almost perfect agreement is obtained for the slightly modified estimation described below. In summary, we have shown that the elastic silver wire/PDMS grating is, indeed, a tunable SPP system.

IV. PROPAGATION AND LOCALIZATION OF SURFACE PLASMON POLARITONS

The focus up to now was the possibility of tuning SPP resonance angles via the grating period. However, the poly-

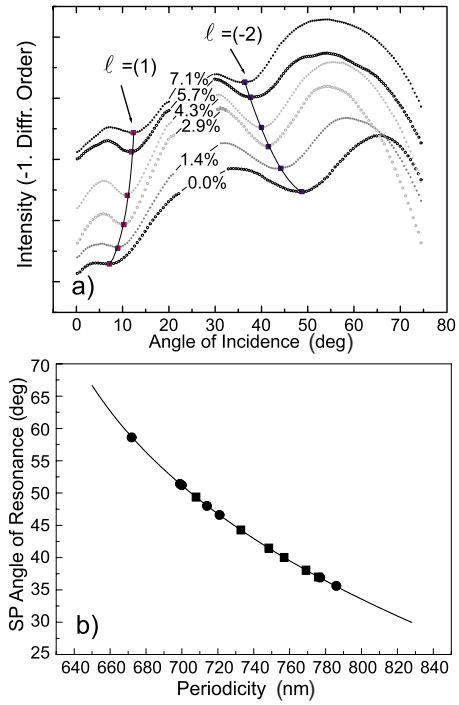

FIG. 3. (Color online) (a) Angular scan of the reflectivity of the tunable system for six different lattice constants, recorded for the −1 diffractive order. (b) Development for the (−2) plasmonic order. The solid line represents a prediction of the plasmon excitation, which proves to be accurate within the limits of the experimental errors; squares show data taken from (a), circles from additional measurements.

mer matrix based metallic wire arrays also allow the study of fundamental questions related to the propagation and localization of SPP's during the transition from a cohesive metallic surface with periodic corrugation to an array of separated silver wires with adjustable distance. Of particular interest is whether the properties of the SPP's can be described by the response of individual wires or whether collective effects involving all wires play the major role. The latter picture underlies the SPP excitation condition given in Eq. (1). However, this equation only defines the possible resonance angles but does not predict the actual magnitude and sharpness of the SPP resonances.

The width and depth of the SPP resonance are mainly determined by the coupling of the SPP to the radiation field. This coupling increases with the spatial variation of the dielectric constant ϵ along the SPP propagation direction.[20,21] It is proportional to the ratio of the spacing between the wires and the period of the grating. Thus, larger slits are expected to cause deeper and broader minima. This trend is, indeed, corroborated by our data, namely by the increasing depth of the minima for stretching up to about 5% [see Fig. 3(a)]. A localization of the plasmon excitation is expected for very large slit widths. This would cause clear deviations from the resonance conditions which are, however, not found in our experiment. We conclude that SPP localization does not occur for the investigated range of wire spacings.

The dependence of the measured minima positions on the grating constant d can be understood in a rather simple pic-

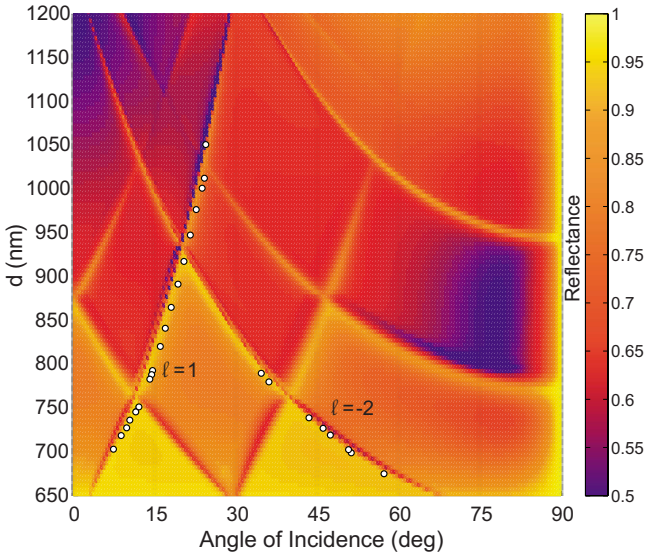

FIG. 4. (Color online) Simulation of the specular reflection of a tunable PDMS based grating with initial periodicity of 640 nm and 40 nm silver thickness. The excitation wavelength is 632.8 nm. The dots indicate the experimentally measured SPP excitations from three different samples.

ture: To account for the finite slit width, the dielectric constant of the metal ϵ_m is replaced by a spatially averaged effective dielectric constant ϵ_{av} (which depends on the dielectric constants of the metal and the polymer, and on the period to slit width ratio) in Eq. (1). Figure 3(b) shows that this yields a remarkably good description.

However, this approach is not sufficient to describe details such as the SPP lifetime as a function of the slit width and the period. Consequently, we have performed simulations of metal wires with variable polymer slits between them using a theory developed by Lochbihler and Depine,[22] which is applicable provided that the conductivity of the metal is very high. Above and below the grating, the electromagnetic field is expanded in plane waves. The metallic character of the wire surface is accounted for by the surface impedance boundary conditions. Within the slits, the field is expanded in slit eigenmodes. The continuity conditions at the grating interfaces determine the field amplitudes and, finally, via Maxwell equations, the electric and magnetic fields above or below the grating. Note that the finite slit width, finite height of the grating, and the dielectric constants of the different media (air and PDMS) at both sides of the grating are taken into account explicitly.

Figure 4 shows calculated specular reflectance for systems with an initial periodicity of 640 nm and stretching up to 1200 nm. A rich SPP "band structure" $d(\alpha_{inc})$ and the formation of gaps in the vicinity of crossings of two respective plasmon modes can be seen. The calculations can be compared directly with the resonance angle measurements, which have been added to Fig. 4 as black dots. Theory and experiment agree within the experimental error, so that we may conclude that our variable spacing system is fully predictable and, therefore, permits the excitation of SPP's for any given wavelength within the tuning range of the system.

Combining theory and experiment opens the possibility to excite and investigate SPP's with special, selected properties.

V. CONCLUSION

This paper clearly shows that excitations of SPP's in different orders can be achieved on the described tunable, smart materials made from metal-coated polymer substrates. Our design allows for the fine control of the lattice constant by simple mechanical means. It was shown that we can adjust the angle of incidence for SPP excitation continuously. This allows one to choose a special point in the band structure $d(\alpha_{inc})$ to excite SPP's and, therefore, to select SPP's with certain desired properties. We have performed simulations that predict SPP properties such as lifetime, group velocity, and emission characteristics at certain combinations of the grating period and the angle of incidence. Combining theory and experiments on tunable lattices allows the potential to study fundamental questions concerning the light-plasmon coupling (i.e., SPP formation) as well as basic properties of SPP's on periodic structures. We found, that simulations compare very well with resonance angle measurements, which is the precondition for subsequent, more detailed investigations.

On a separate viewpoint, the presented tunable systems allow for the design of previously unknown types of spectroscopic methods. The ability to tune the wavelength of the exciting photons while maintaining SPP excitation results in extreme surface sensitive and selective variants of major techniques. The spatial confinement of the generated optical near field and the SPP sensitivity are further arguments along this line. Due to the inexpensive production of the tunable chips, they can also be targeted for a mass production market, for example, in the life sciences.

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