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IEEE Photonics Journal
An IEEE Photonics Society Publication

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Volume 6, Number 5, October 2014

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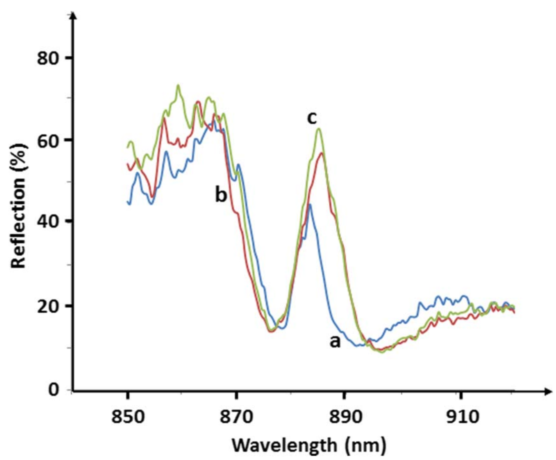

DOI: 10.1109/JPHOT.2014.2361664

Sharp Plasmon-Mediated Resonant Reflection From an Undulated Metal Layer

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DOI: 10.1109/JPHOT.2014.2361664

Manuscript received August 9, 2014; revised September 24, 2014; accepted September 26, 2014. Date of current version October 20, 2014. Corresponding author: Y. Jourlin (e-mail: yves.jourlin@univ-st-etienne.fr).

Abstract: A close-to-the-theoretically-largest TM resonant reflection of a free-space wave from a free-standing undulated gold layer immersed in a liquid host medium is demonstrated experimentally. It is mediated by the grating-excited long-range plasmon mode propagating along the continuous metal film with particularly low loss.

Index Terms: Gratings, plasmonics, long-range plasmon, resonant reflection, metal film.

1. Introduction

It is known from the mid 1980's that the spectrally sharp and theoretically 100% resonant reflection of a free-space wave from a dielectric grating waveguide is related to the excitation of a waveguide mode [1]. This is usually understood to be destructive self-interference in the transmission medium between the field coupled into the waveguide and then re-radiated to the adjacent media, and the field of the transmitted 0th order. In a lossless symmetric dielectric waveguide there is a 100% transmission peak close to the 100% reflection peak. This reflection effect can be easily observed and has been used in a number of fields of application such as biosensors [2] and counterfeit deterrence [3], among others. A similar resonant reflection effect should in principle also take place in an undulated metallic structure propagating a surface wave of the plasmonic type. However, there are two major differences with the dielectric grating waveguide case: firstly, there is no transmission medium, and secondly, the propagation loss of the plasmon mode at a single interface is usually so high, i.e., its propagation length is so short (a few microns at optical frequencies), that it is not possible for a corrugated grating to produce the radiation strength needed for the coupled surface plasmon to be radiated out from the waveguide before it is absorbed. The situation changes radically if, instead of a thick substrate, the metal has the form of a continuous thin film whose thickness is of the order of the penetration depth of the field: in this case, there is a transmission medium and, if the film is embedded in a homogenous host medium, the so-called long-range plasmon mode has particularly low propagation losses since its electron-driving longitudinal electric field has a zero-crossing in the middle of the metal film. This mode has been used in low-loss plasmonic integrated optical elements operated at telecommunication wavelengths [4]. Under such conditions, the coupling and re-radiation mechanism applies, since a shallow undulation of the metal layer can have a radiation coefficient greater than the absorption coefficient of the long-range plasmon. However,

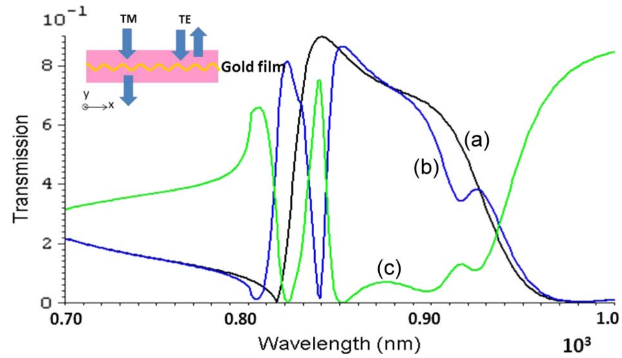

Fig. 1. Theoretical TM spectra of a sinusoidally undulated gold film. (a) Transmission at normal incidence. (b) Transmission at a 1° incidence angle. (c) Reflection associated with transmission curve (b).

of the two neighboring reflection and transmission resonance peaks, the latter has been so far observed experimentally in the form of a broadband tunneling of light through the continuous metal film [5], mediated by two entangled, short- and long-range plasmon modes (the film was used as a polarizer [6]), whereas the resonant reflection peak overlaps with the edge of the strong, broad, non-resonant metallic reflection spectrum of the metallic film, and can thus not be resolved. Whether resonant plasmonic reflection exists has long been an open issue [7]. It is by the way a rather counterintuitive issue since, as it is well known, resonant plasmon coupling by a grating usually results in a broad and deep reflection dip, not in a sharp reflection peak. The existence of a reflection peak was only very recently shown in a quasi-symmetrical structure where an undulated silver film was deposited onto a sinusoidal photoresist grating, covered by a superstrate having close to the same refractive index as the resist with two identical dielectric interfacial nanolayers (ZnS and MgF_2) to avoid index inhomogeneities and local delaminations at the metal surfaces [8].

The object of the present paper is to provide the experimental evidence of the existence of this sharp TM reflection phenomenon under conditions where the metal film is perfectly embedded in its host medium without any interfacial loss problem. Such situation occurs if the host medium is a liquid surrounding a free-floating undulated metal layer. Resonant reflection at a metal surface is however difficult to observe, since the surface is highly reflective even outside resonance. A further difficulty arises from the fact that the long-range plasmon is very sensitive to index and adhesion inhomogeneities at the dielectric-metal interfaces, where its field strength is at a maximum [6]. It thus suffers from considerable excess losses, which degrade the resonant effect: although more than 90% resonant transmission is expectable in theory through an undulated continuous gold layer in the near-IR, a maximum TM transmission of no more than 50–60% was measured. Thus, carefully designed necessary experimental conditions must be created for resonant reflection to be observable and measurable. What makes the experimental evidence of resonant reflection difficult also renders practical applications of the effect problematic which requires the understanding of its nature to make use of its polarization and spectral properties.

2. Experiment Rationale

The first condition for an observable reflection peak, if it exists, is to experimentally provide a spectrally wideband dark-field reflection background, on which the sought reflection peak would be comparatively bright. This condition was fulfilled by using a periodic undulation (a grating) to couple an essentially normally incident wave to the long- and short-range plasmons of a symmetrically embedded, continuous metal film exhibiting the spectrally wide transmission peak of the TM polarization [6]. This experimental condition was modeled exactly using the Chandezon method [9] on a typical sample, as shown in Fig. 1, with a continuous, 30 nm thick gold film, characterized by a double-sided sinusoidal undulation having a period equal to 540 nm and a depth equal to 70 nm, embedded in a homogeneous medium of index 1.5. In the inset of Fig. 1 the x-axis is along the grating K-vector, the y-axis is parallel to the grating lines; the TM polarization has its magnetic field H along y whereas the electric field E is along y in the TE polarization. The TM transmission spectrum shown in curve (a), obtained with the complex permittivity of bulk gold, has a peak transmission of approximately 90%, and a shoulder at a longer wavelength. The latter are the resonant transmission contributions of the long- and short-range plasmons, respectively, the short-range plasmon having a higher effective index n_e and a larger absorption loss. Under normal incidence the relationship between the incident wavelength λ and the grating period Λ coupling the incident wave to a plasmon mode of effective index n_e is $\Lambda = \lambda/n_e$. n_e in a thin metal film embedded in a uniform host medium of index n has two values determined by a transcendental dispersion equation containing the metal layer thickness relative to the incident wavelength and the permittivities of the metal and adjacent medium. In [8], the dispersion equation of the most important (because of its low propagation losses) long-range plasmon mode is given.

It is important to note that the two resonant transmission features (peak and shoulder due to the long-range and short-range plasmon modes, resp.) are entangled. There is no fall in transmission between these two resonances, even when the imaginary part of the metal permittivity is artificially and arbitrarily decreased; this is the consequence of the strong coupling between the two plasmon modes, provoked by a conformal undulation on both sides of the metal film [7]. Curve (a) in Fig. 1 shows a transmission zero located to the left of the long-range transmission peak: it is where a resonant reflection peak could be found if it exists. However, this zero is in the form of an edge with sharp increase at one side only. From this, it is not possible to determine firstly, whether the transmission zero corresponds to a reflection and/or an absorption effect, and secondly, whether there is a peak hidden by this edge. The designed experimental artifice is to slightly tilt the incidence off-normal in a plane normal to the grating lines to remove the degeneracy between the $+$ and -1st grating orders. As shown in curve (b) of Fig. 1, this has the effect of shifting the transmission zero towards the resonant transmission peak of the $+1$st order, where it now appears as a sharp, well-defined transmission notch in a spectrally broad high transmission band. The reflection spectrum plotted in curve (c), associated with the transmission spectrum of curve (b), shows that this notch is indeed associated with a nearly 80% sharp reflection peak, with a spectral width equal to 10 nm, and with a moderate increase in absorption, confirming the mediation of a resonant plasmonic effect: the absorption loss in Fig. 1 is given by 1 minus the sum of curves (b) and (c); as expected the largest losses are at the side of the short-range plasmon (right-hand side of the spectrum), but there is also a local loss maximum of 26% right at the reflection peak.

For the resonant reflection peak to be observable, if it exists, the second necessary condition which must be satisfied is the avoidance of excess plasmon losses, which can take place at the metal/dielectric interfaces, characterized by non-uniform adhesion and local overheating of the polymer when the first drops of molten metal impinge on its surface, as reported in [10]. The experimental model of an ideal structure, shown in the inset of Fig. 1, is comprised of a glass substrate underlying a ca. 1 μm thick photoresist layer with a sinusoidal profile grating, coated with a thin gold film. The undulated metal film is then coated by an optically thick photoresist layer, which provides the dielectric host medium with optically symmetrical properties, ensuring guidance of the long-range plasmon mode. An experimental structure was fabricated with a 30 nm thick gold film, and an ample undulation having a period equal to 600 nm and a depth of 140 nm nominally, and actually between about 100 and 140 nm after the release of the gold film from the undulated resist substrate; it is not identical to the modeled structure, however, the main features of the transmission/reflection mechanism are preserved. In order to suppress the excess losses of the propagating plasmon modes, the interface inhomogeneity problem was removed by dissolving the adjacent photoresist layers. The element was installed horizontally in a bowl filled with acetone, in a vertically oriented transmission spectroscopy mount allowing the transmission spectrum to be acquired while the resist was dissolving. The results of the transmission

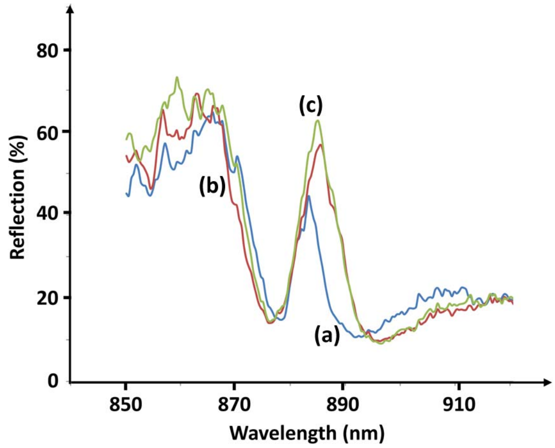

Fig. 2. Measured TM reflection spectra of an immersed gold grating under 1° incidence. Curves (a)–(c) correspond to increasing time during the resist dissolution process.

measurement are amply described in [9]; the measured transmission spectrum was close to the theoretical transmission spectrum of curve (b) in Fig. 1, which exhibits under 1 degree incidence a sharp dip in the broad transmission maximum and therefore provides the conditions of observation of a possible reflection peak on a dark reflection background as the next section will show. The reason for which the transmission peak is so broad is not related to the plasmonic losses, but to the fact that this type of double conformal undulation strongly couples the long- and short-range plasmons [7].

3. Evidence of Sharp Plasmon-Mediated Resonant Reflection

Fig. 2 shows the measured reflection spectrum to be compared with the theoretical reflection spectrum (c) of Fig. 1. It was obtained by means of a beam splitter placed in the path of the quasi-normal incident TM beam (H field along the y-axis) at about 1 degree incidence. It clearly reveals the presence of a sharp resonant reflection peak at 885 nm. It shows three curves of increasing peak height that were recorded at different times while the photoresist was dissolving. Although most of the photoresist was dissolved after a few minutes, leading to the appearance of a reflection maximum (curve (a)), it took more than one hour to dissolve the large number of bridging points between the undulated gold film and the overheated resist clusters created at the very beginning of the gold evaporation. The measured spectra (curves (b) and (c)) reveal the presence of slow instabilities as the bridging points give away (ATG instabilities [11]) to finally reach a stable maximum (curve (c)). These instabilities are the effect of the non-uniform separation in the fluid of the gold layer from its polymer substrate due to local stronger adhesion and smaller resist solubility at the overheated points. The spectra of Fig. 2 are expressed in % of the reflection of a non-immersed plane gold surface as the reference (the reflection reference was nearly constant at 98% over the rather narrow measurement spectrum of 80 nm). This peak is found to correspond to a reflection of ca. 60%, since it is approximately 10% lower than the 70% reflection maximum located at 860 nm at the left edge of the left reflection dip. The dips at either side of the reflection peak are non-zero, which is attributed to the 4% Fresnel reflection at the air/water and bottom interfaces. The spectral width of the reflection peak is about 10 nm, i.e., ca. 10 times narrower than the overall transmission peak. The reflection peak is also ca. 3 times narrower than the plasmon-excitation dip in the reflection spectrum of an infinite gold substrate having the same surface undulation. Whereas the broad spectrum of resonant transmission is clearly the result of the contribution of both long-range and short-range plasmon modes [10] as shown by curve (a) in Fig. 1, it is remarkable that the narrow spectrum of resonant reflection is the result of the sole long-range mode effect.

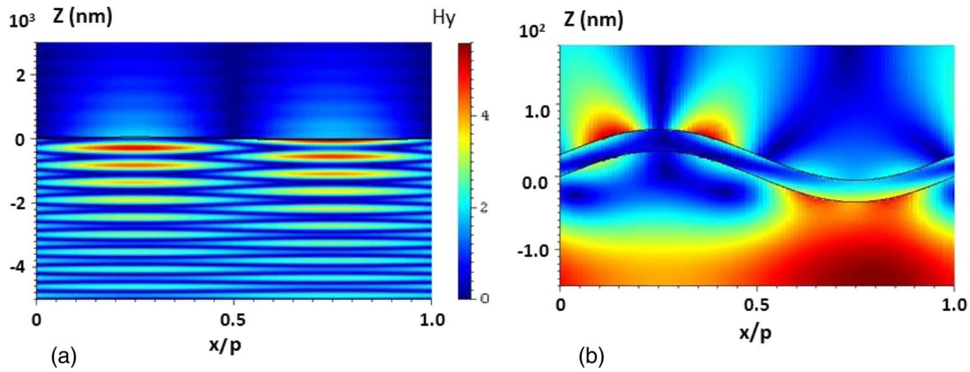

Fig. 3. TM field maps at resonance over one grating period along x. (a) Transverse magnetic field at normal incidence (see curve (a)) of Fig. 1, 816 nm wavelength. (b) Zoomed longitudinal electric field along x at 1° incidence angle (see curves (b) and (c) of Fig. 1), 837 nm wavelength.

This experiment demonstrates the existence of resonant reflection from an undulated metal layer embedded in a homogeneous host medium, and shows that its modulus is close to that computed using the complex permittivity of the bulk material, even though the layer of gold was not deposited by the slow rate evaporation process recommended for low loss plasmonic waveguide layers.

The statement that the reflection peak (and the associated zero transmission notch) corresponds to plasmonic resonance is supported by the false color field map, taken over one grating period along x, shown in Fig. 3(a)).

It represents the transverse magnetic field distribution H(x) corresponding to the zero transmission (curve (a) of Fig. 1) at normal incidence and a wavelength of 816 nm. The scan along the normal to the metal film was intentionally extended far from the latter, to allow the regular and resonant components of the field to be clearly distinguished: in the transmission half-space (top of the field map), far from the undulated metal film, the field tends to zero, whereas in the incidence half-space (bottom), it has a standing wave distribution corresponding to a planar reflected wavefront, the standing wave corresponding to the superposition of the incident and reflected waves. The field is clearly concentrated in the region of the film, which confirms the mediation of a plasmon resonance. The resonant part of the field, which has the same symmetry as the film undulation, fades away rather slowly in both adjacent half-spaces, because the effective index of the long-range plasmon is very close to the index of the host medium, thus its exponentially decreasing field tail extends far away from the metal layer.

Fig. 3(b) is a zoomed map of the longitudinal electric field, which is mostly responsible for the driving of the free electrons in the metal, therefore of the absorption losses (as in Fig. 3(a)), the top (bottom) half-space is the transmission (incidence/reflection) medium). The incidence angle is here 1 degree to place the resonant reflection in the dark field created by the broad resonant transmission spectrum as it was set up experimentally for obtaining the measured reflection spectrum of Fig. 2. As expected, this electric field component exhibits zero crossings within the metal film, which is typical for the long-range plasmon mode. The resonant reflection of curve (c), Fig. 1, under 1 degree incidence, occurs at the wavelength of 837 nm; the field distribution is slightly asymmetrical as a consequence of the non-normal incidence angle.

4. Conclusion

In conclusion, the present study shows that a continuous, thin undulated metal layer embedded in a homogeneous host medium exhibits resonant reflection, similar to the known resonant reflection observed in a corrugated dielectric slab waveguide, and that its high reflection modulus is mediated by the long range plasmon. This demonstration is an interesting scientific result in itself. Its potential application as a mirror is however limited by the inherent absorption losses, which is the case for most plasmonic effects. Nevertheless, the fact that so narrow a spectrum is produced by so simple a structure is potentially interesting as a basis mechanism applicable for security features. Besides, an interesting feature of such an element is the association of wavelength and polarization reflection selectivity with electrical conductivity, which could be useful in an electrically pumped semiconductor and organic light emitters. Such perspectives should be evaluated at the light of the abundant research work made in the domain of surface plasmon-polariton light emission and amplification [12]. The resonant reflection effect was demonstrated here with a liquid host medium which of very limited applicability. There is however some hope to achieve comparably low losses with solid interfaces with the BCB-gold-BCB plasmonic waveguide system used successfully in the telecom C-band [4].

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