A surface plasmon resonance spectrometer using a super-period metal nanohole array

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Abstract: We investigate the surface plasmon resonance in super-period nanohole arrays and demonstrate a surface plasmon resonance spectrometer using a super-period metal nanohole array device. Super-period nanohole arrays are patterned metal nanohole array gratings. In a super-period nanohole array, there is a small subwavelength nanohole period that supports local surface plasmon resonance, and also a large grating period that diffracts surface plasmon radiations to non-zeroth order diffractions. With the super-period metal nanohole array, surface plasmon resonance can be measured in the first order diffraction in addition to be traditionally measured in the zeroth order transmission. The resonance peak wavelength measured in the first order diffraction is slightly blue-shifted from the resonance wavelength measured in the zeroth order transmission.

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OCIS codes: (250.5403) Plasmonics; (300.6190) Spectrometer.

References and links

1. Introduction

Extraordinary optical transmission through periodic nanohole arrays in opaque metal films was first reported in 1998 [1]. The phenomenon has been extensively investigated for more than a decade [2–11]. Enhanced light transmission through subwavelength period nanohole arrays occurs when the frequency of incident light is tuned to the plasmon resonance frequency of the periodic metal nanohole arrays. The coupling between the surface plasmon Bloch wave and the nanohole dipole local plasmon resonance contributes to the enhanced light transmission through the nanohole structured metal film.

Extraordinary optical transmission phenomenon and the underlined surface plasmon resonance can only be measured in either the transmission or the reflection because the period of the nanoholes in the regular array is subwavelength. In this paper, we propose a new metal nanohole array structure with which the surface plasmon resonance can be measured differently. The new nanohole array structure has two periods: one is a small subwavelength period that enhances surface plasmon resonance and light transmission. Another period, called “super-period,” is created by periodically removing lines of the nanoholes in regular periodic nanohole arrays. The super-period functions as a grating period which is above the wavelength of interest. Because of the super-period structure, surface plasmon radiations from metal nanohole arrays can be measured from the non-zeroth order diffractions. Due to the intrinsic angular dispersion of diffractions, the resonance spectrum in non-zeroth order diffractions can be measured with a linear photodetector array or a CCD. Recently, we reported a surface plasmon resonance spectral sensor using a super-period metal nanoslit array device [12] and also predicted that the first order diffraction resonance is more close to the near field resonance than the zeroth order transmission resonance [13]. In this work, we experimentally demonstrate a surface plasmon resonance spectrometer using a one dimensionally (1D) patterned two dimensional (2D) nanoholes in a thin gold film. Also we find that the first order diffraction resonance peak wavelength is slightly blue-shifted from the zeroth order transmission resonance peak wavelength.

2. Design and simulations

A super-period nanohole array is a one dimensional diffraction grating of two dimensional nanohole arrays with a small nanohole array period p and a large grating period P, as illustrated in Fig. 1(a). The large grating period P is five times of the small period p in our first design. The super-period nanoholes have a small period of 420 nm and a super grating period of 2100 nm in a gold film of 50 nm thick on a quartz substrate. The zeroth order light transmission and the first order diffraction from the device were calculated with the normal light incidence. The polarization of the incident light is along the effective nanohole array grating lines, which is normal to the direction of diffractions. Calculations were carried out using a finite difference time domain (FDTD) software code developed by Lumerical Solutions, Inc. Figure 1(b) shows the calculated zero-order transmission spectrum (the black dashed line curve) and the first order diffraction spectrum (the solid red line curve) from the super-period nanohole array device. It can be seen that two plasmon resonance modes are excited in the device. The resonance at the longer wavelength corresponds to the tightly confined surface plasmon mode that strongly couples the local oscillations of the nanohole dipoles. This plasmon resonance mode is also called the “substrate mode” because it provides strong electromagnetic field enhancement in the metal-substrate interface. The resonance at the shorter wavelength corresponds to the weakly confined surface plasmon mode. This mode is also called the “air mode” because it provides strong electromagnetic field enhancement in the metal-air boundary. In Fig. 1(b), it can be seen that the zeroth order transmission peak due to the tightly confined plasmon resonance mode is at 760.5 nm wavelength and the first order diffraction peak due to the same plasmon resonance mode is at 750.5 nm wavelength.
From the calculated transmission and diffraction in Fig. 1(b), it can be seen that the surface plasmon resonance in the super-period nanohole array can be observed from the first order diffraction and also from the zeroth order transmission. However, resonance peak wavelength in the first order diffraction is 10 nm blue-shifted from the peak wavelength in the zeroth order transmission. To gain further information on the resonance, we calculated the electric field intensity distribution on a plane 20 nm above nanohole metal surface at 750.5 nm and 760.5 nm respectively, and plot the calculation results in Figs. 2(a) and 2(b). It is seen that the electric field is stronger at the 750.5 nm wavelength than the electric field at 760.5 nm wavelength on the near field plane 20 nm above the metal nanohole surface. We also calculated the electric field intensity versus the wavelength at the top center location of one of the two inner nanohole apertures within a super-period unit cell. The result is plotted in Fig. 3(a). It can be seen that the strongest field enhancement at this location is at 750.5 nm wavelength. We also calculated the electric field intensity versus the wavelength at the top center of one of the two outer nanohole apertures within a super-period unit cell and plotted the calculation result in Fig. 3(b). The strongest field enhancement is at 749.5 nm wavelength at this location. Here we see that the first order diffraction peak wavelength of 750.5 nm is approximately the same as the near field resonance wavelength although the near field resonance wavelength slightly depends on the location of measurement.
radiations and the directly transmitted light through the nanoholes and the thin metal film. For a 50 nm gold film, a significant amount of light can transmit through the film. Near the nanohole structured metal film, the surface plasmon resonance enhanced near field is strong and dominant. Therefore, the near field resonance wavelength is primarily determined by the surface plasmon resonance. The far field diffractions, either the first order or higher orders, avoid the interference between the surface plasmon radiations and the transmission. Therefore, the resonance in diffractions is directly related to the near field resonance. It is known that in metal nanoparticles, the near field resonance occurs at longer wavelength than the resonance peak wavelength in the scattering and the peak wavelength in the absorption due to the damping of the surface plasmon oscillation [14–17]. For the super-period nanohole array, we also calculated the overall absorption versus the wavelength. We find that the absorption in the device reaches the maximum at 743.5 nm wavelength, which is blue-shifted from the near field resonance wavelength. The blue shift of the peak absorption wavelength from the near field resonance wavelength is consistent with the report in [14–17].

3. Experimental results

A super-periodic nanohole array device was fabricated in a 50 nm thick gold film on a glass substrate with a standard e-beam lithography patterning and argon ion etching process. The SEM picture of the e-beam lithography patterned super-period metal nanohole device is shown in Fig. 4. The diameter of the nanoholes in the array is approximately 140 nm. The small period of the nanoholes is 420 nm. The super-period is 2100 nm. The total patterned nanohole area is 300x300 µm².

The super-period nanohole array device was measured with a super continuum broadband laser source (from NKT Photonics, Inc.). The excitation light was normally incident from the substrate with the polarization parallel to the nanohole effective grating lines. A CCD imager (Sony ICX098BQ) was used to capture the angularly dispersed intensity distribution of the first order diffraction from the nanohole patterned grating. Figure 5 shows the color coded images of the spatially dispersed first order diffraction when different liquid chemicals were applied to the device surface area. To obtain the first order diffraction spectrum, a calibration is needed to obtain the correspondence between the CCD pixels and the wavelengths. We use a Helium-Neon (HeNe) laser of 632.8 nm wavelength to calibrate the measurement setup. The HeNe laser is aligned to propagate in the same direction as the broadband laser. We first identify the pixel that corresponds to 632.8 nm wavelength on the CCD. Once the pixel corresponding to 632.8 nm wavelength is known, the correspondence between all pixels on the CCD and wavelengths can be obtained by using the diffraction equation.
\[
\sin(\theta) = \frac{x}{\sqrt{d^2 + x^2}} = \frac{\lambda}{P},
\]

where \( \theta \) is the first order diffraction angle, \( x \) is the distance between the first order diffraction spot and the zeroth order transmission spot on the CCD plane, \( d \) is the distance between the device and the CCD, \( P \) is the super grating period, \( \lambda \) is the free space wavelength corresponding to \( x \). The distance \( d \) is 14.8 mm in our experiment setup. After the calibration, the first order diffraction can be obtained by normalizing the CCD signal with the responsivity of the CCD photodetector pixels. Adjustments were made to correct for the small beam misalignment between the surface normal and the incidence.

Fig. 4. A SEM picture of the e-beam patterned super-period nanohole array in a thin gold film.

Figure 5(a) is the color coded, angularly dispersed first order diffraction image (diffracted along the horizontal axis) captured by the CCD when the device is in the air. Figure 5(b) is the color coded, spatially dispersed first order diffraction image captured by the CCD when methanol liquid is applied to the device surface. Figure 5(c) is the color coded, spatially dispersed first order diffraction image when isopropyl-alcohol is applied to the device surface. The images are the intensity signals captured by the CCD. The indexes of refraction of methanol and IPA liquids are 1.328 and 1.375, respectively.

Fig. 5. Spatially dispersed first order diffraction images captured by a CCD when the super-period metal nanohole array device area was exposed to: (a) the air, (b) the methanol, and (c) the isopropyl-alcohol.

Figure 6(a) shows the zeroth order transmission spectra when different liquid chemicals are applied to the nanohole device surface. The resonance peak wavelength of the stronger resonance at the longer wavelength in the zeroth order transmission is 790 nm when the device exposes to the air. The resonance peak wavelength shifts from 790 nm to 804 nm when methanol is applied, and shifts again to 814 nm when isopropyl alcohol (IPA) is applied. The sensitivity for the index of refraction is 213 nm/RIU based on the zeroth order transmission measurements. Figure 6(b) shows the first order diffraction spectra when different liquid
chemicals are applied to the device. The spectra in Fig. 6(b) are measured by the CCD and normalized to the spectrum of incident broadband light source. The vertical axis in Fig. 6(b) has an arbitrary unit. It can be seen that the resonance wavelengths in the first order diffraction are slightly blue-shifted from the resonance peak wavelengths in the zeroth order transmission. When chemicals are applied to the device surface, the peak diffraction wavelengths in the first order diffraction are shifted. Tracking the shift of the diffraction peak wavelength of the longer wavelength resonance, it is found that the first order diffraction peak wavelength shifts from 778 nm in the air to 794 nm in the methanol, and again shifts to 809 nm in the IPA. By calculating the sensitivity based on the first order diffraction peak wavelength shift from the methanol to the IPA, the sensitivity for index of refraction is 319 nm/RIU.

![Fig. 6. Measured surface plasmon resonance spectra when different liquid chemicals are applied to the super-period gold nanohole array surface: (a) the zeroth order transmission spectra measured by using a commercial spectrometer and (b) the first order diffraction spectra obtained by using our integrated surface plasmon resonance spectrometer.](image)

4. Summary

Surface plasmon resonance in super-period metal nanohole arrays are investigated by calculating resonance spectra of the zeroth order transmission, the first order diffraction, and the near field resonance. It is found that the first order diffraction resonance peak wavelength is slightly blue-shifted from the zeroth order transmission peak wavelength. It is also found that the first order diffraction peak wavelength is approximately the same as the near field resonance wavelength although near field resonance wavelength slightly varies with the location of measurements. An integrated surface plasmon resonance spectrometer based on an e-beam lithography patterned super-period metal nanohole array is experimentally demonstrated. The surface plasmon resonance spectrometer can measure surface plasmon resonance from the spatially dispersed first order diffraction with a single shot CCD data image capture. The integrated surface plasmon resonance spectrometer based on the super-period metal nanohole array performs the functions of surface plasmon resonance sensing and resonance spectral measurements simultaneously.

Acknowledgments

This work was partially sponsored by the National Aeronautics and Space Administration (NASA) through the grants NNX07 AL52A and NNX12AI09A, and partially sponsored by the National Science Foundation (NSF) through the award NSF-0814103.