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Abstract. The excitation of multiple surface-plasmon-polariton (SPP) waves and waveguide modes was experimentally demonstrated in a structure fabricated as a one-dimensional photonic crystal (PC) on top of a two-dimensional metal grating. The PC had two periods, each period comprising nine layers of silicon oxynitrides of different compositions. The individual excitations of the SPP waves and waveguide modes were also theoretically predicted using the Floquet theory, surface-multiplasmonics theory, and the transfer–matrix approach for multilayered waveguides. Both the theoretical and experimental results indicate broadband coupling of incident light of either linear polarization state to the guided waves of both types over a broad range of the angle of incidence. © *2015 Society of Photo-Optical Instrumentation Engineers (SPIE)* [DOI: 10.1117/1.JNP.9.093593]

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1 Introduction

Surface-plasmon-polariton (SPP) waves are electromagnetic surface waves guided by the planar interface of a dielectric material and a metal.[1] These waves are commonly exploited for optical sensing[2] and biosensing,[3] optical filtering,[4] and photo detection.[5] Of particular interest, in recent years, has been their application for photovoltaic[6] and photoelectrochemical[7] energy-harvesting devices, because their excitation can offer light-absorption enhancement exceeding the Lambertian limit.[8]

Although absorption enhancement by SPP waves in photovoltaic devices has been studied for more than three decades,[9–12] significant improvements in device performance have not been reported yet. This is because the SPP waves can only enhance the absorption by a small amount—since (1) only incident *p*-polarized light (magnetic field perpendicular to the incidence plane) can excite SPP waves and (2) only one SPP wave can be excited at a specific free-space wavelength when the dielectric material is homogeneously normal to the

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interface.[13] In addition, the field is confined in a subwavelength region near the interface, leading to high absorption in the metal and limited enhancement in the active device region that is usually quite far from the interface.[12]

When the dielectric material is of finite thickness, the air/dielectric/metal structure can act as an open-face slab waveguide.[14,15] The fields of the waveguide modes are largely confined to the dielectric slab and depend on the thickness of that slab. These waveguide modes can also enhance the electric field and, therefore, the electron–hole pair generation rate, in photovoltaic solar cells.[16–18]

When the dielectric material is periodically nonhomogeneous in the thickness direction, multiple SPP waves can be guided by its interface with the metal.[13,19] The excitation of the multiple SPP waves has been experimentally confirmed [20–22] over a broad range of incidence angles and wavelengths. Furthermore, the underlying surface-multiplasmonics theory[19] shows that light absorption in thin-film solar cells can be increased by introducing periodic nonhomogeneity in the semiconductor material.[23] Finally, because the propagation length of some of the multiple SPP waves in this structure is predicted to be as long as millimeters, the interface can be potentially exploited as a planar solar concentrator.[24]

Most often, experimental excitation of SPP waves requires either the use of a coupling prism or the periodic corrugation of the metal/dielectric interface.[25] For a solar-energy-harvesting application, the use of a prism[9] is impractical but the periodic corrugation of the interface has been investigated for three decades[10,11] and is very practical. Of course, these investigations were confined to the traditional case of optically homogeneous semiconductors.

In order to experimentally verify the impact of surface multiplasmonics on light absorption, we first deposited a periodically multilayered material—i.e., a one-dimensional (1-D) periodic crystal (PC)—on a 1-D metallic grating and then measured the specular reflectance of this structure over broad ranges of the angle of incidence θ (with respect to the thickness direction) and the free-space wavelength λ_0.[22] The incidence plane was made to coincide with the grating plane in the optical experiments. Our experimental findings were consistent with theoretical predictions. Most importantly, we confirmed that, at a fixed value of λ_0, the same SPP wave can be excited as Floquet harmonics of different orders by light incident at different values of θ. Waveguide modes can also be excited as prolifically.[16,18,24]

If the 1-D grating were to be replaced by a 2-D grating, many more Floquet harmonics would exist,[26] leading to increased possibilities of exciting multiple SPP waves and waveguide modes. Accordingly, guided-wave propagation could occur in several directions that are not restricted to lie in the incidence plane. Although the theoretical formulation to obtain rigorous solutions exists,[Ref. 19, Sec. 3.8], it requires computational resources that are still not easily available. Fortunately, in order to analyze the results of corresponding experiments, full-scale theory is nonessential and just a simple consequence of Floquet theory suffices. This has been demonstrated for multiple SPP waves when 1-D[22] gratings are used.

In this paper, we experimentally demonstrate the excitation of the multiple SPP waves and waveguide modes with a structure constructed as a 1-D PC of finite thickness on top of a 2-D metal grating. To our knowledge, this is the first experimental domenstration of the excitation of multiple SPP waves with 2-D gratings.

The plan of this paper is as follows. Experimental work is described in Sec. 2. Surface-multiplasmonics theory to calculate the SPP wavenumbers is presented in Sec. 3.1, the transfer–matrix approach to determine the wavenumbers of waveguide modes is provided in Sec. 3.2, and predictions for the experimental observation of SPP waves and waveguide modes are discussed in Sec. 3.3. The obtained experimental results are presented and compared with the predictions in Sec. 4. The paper ends with final remarks in Sec. 5.

2 Description of Experiments

Figure 1(a) provides a schematic of two unit cells in both the x and y directions of the fabricated structure. The grating was made of gold (Au) using a template-stripping process. A silicon wafer was patterned by electron–beam lithography on ZEP520A photoresist (Zeon, Tokyo) with the inverse pattern of the metal grating. The pattern in the photoresist was transferred into the silicon

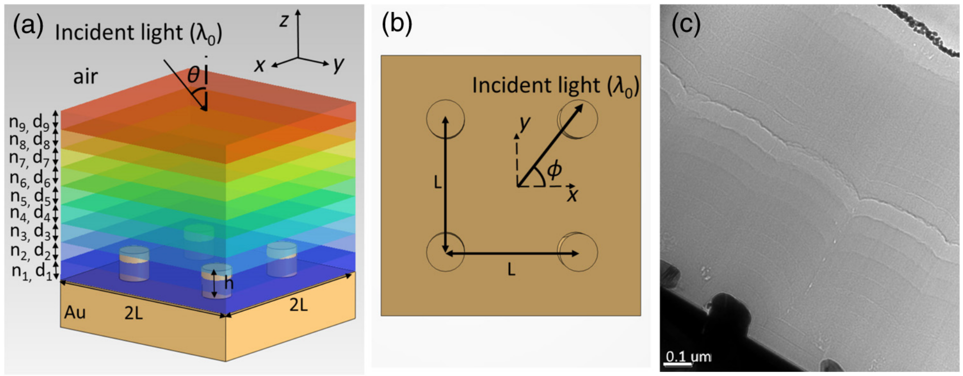

Fig. 1 (a) Schematic of a one-dimensional (1-D) photonic crystal (PC) deposited on top of a two-dimensional (2-D) metal grating. Only one period of the PC is shown. The wavevector of the incident light is inclined at θ with respect to the z axis, and (b) at ϕ with respect to the x axis in the xy plane. (c) Transmission electron microscopy image of the fabricated sample with two periods of PC.

by inductively coupled reactive-ion etching on a Versalock 700 (Plasma-Therm, St. Petersburg, Florida) with pure chlorine gas. The photoresist was dissolved in a commercially available resist remover named nanostrip with the assistance of a bath sonicator. Gold was thermally evaporated on the patterned silicon wafer at room temperature with a base pressure of $<10^{-6}$ Torr at a deposition rate of $0.075\ \mathrm{nm\,s^{-1}}$. The gold film was attached to a glass slide using EpoTek 377 epoxy (Epoxy Technology, Billerica, Massachusetts). The epoxy resin was cured at 100 °C for 7 days. Then, the gold film was released from the silicon wafer with a razor blade, yielding a $6 \times 6\ \mathrm{mm}^2$ gold grating glued to a glass slide.

The grating has a period $L = 350$ nm in both the x and y directions. Each square unit cell of the grating contains a circular step of height of 90 nm and cross-sectional diameter of 190 nm. The complex refractive index of gold, which was calculated from data obtained using an RC2 spectroscopic ellipsometer (Woollam, Lincoln, Nebraska), is presented in Fig. 2.

The PC on top of the 2-D gold grating has two periods, although only one period is shown in the figure. Each period consists of nine silicon oxynitride $(\mathrm{SiO_2})_a(\mathrm{Si_3N_4})_{(1-a)}$ layers of different compositions identified by $a \in [0,1]$. The silicon oxynitride layers were deposited using plasma-enhanced chemical vapor deposition on a cluster tool (Applied Materials, Santa Clara, California) at a susceptor temperature of 220 °C. Ammonia, silane, and nitrous oxide were used in varying ratios to deposit layers with specific refractive indices. All layers were deposited at a pressure of 3.5 Torr and a power density of $0.955\ \mathrm{W\,cm^{-2}}$.

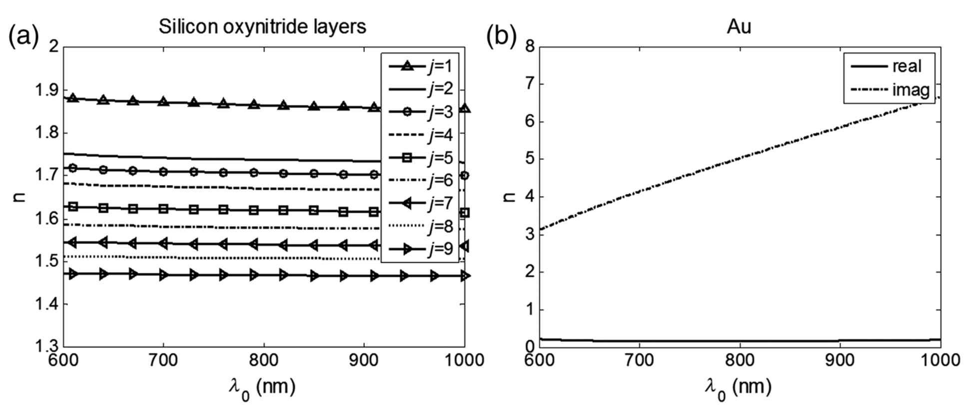

Fig. 2 (a) Spectrums of the refractive indices of the nine silicon-oxynitride layers in each period of the 1-D PC. The imaginary parts of the refractive indices were on the order of 10^{-4} (Ref. 22) and were therefore ignored. (b) Spectrums of the real and imaginary parts of the refractive index of gold.

The refractive index n_j of the j'th layer, $j \in [1, 9]$, was measured in the same way as that of the gold layer. The refractive indices of all nine layers are provided in Fig. 2. The transmission-electron microscopy image shown in Fig. 1(c) was used to measure the thickness d_j of the j'th layer, $j \in [1, 9]$. The measured thicknesses are as follows: $d_1 = 58$, $d_2 = d_5 = d_6 = 60$, $d_3 = 46$, $d_4 = 59$, $d_7 = 67$, $d_8 = 71$, and $d_9 = 78$ nm. Let us note that the compositions of the nine layers were merely chosen for ease of fabrication in our nanofabrication facility, and other materials that are virtually nondissipative in the solar spectral regime could be used instead of silicon oxynitride.

The specular reflectances R_{s0} and R_{p0} of the structure were measured for $\theta \in [8\ \text{deg}, 55\ \text{deg}]$ and $\phi \in \{0\ \text{deg}, 45\ \text{deg}\}$, as shown in Figs. 1(a) and 1(b), on a custom spectrometer for incident light of s and p polarization states, respectively. The measurements were carried for $\lambda_0 \in [600, 1000\ \text{nm}]$. A schematic diagram of the custom spectrometer is presented in Fig. 3. A halogen light source (HL-2000, Ocean Optics, Dunedin, Florida) is connected to an optical fiber ($f1$) (QP400-2-VIS-NIR, Ocean Optics, Dunedin, Florida) which guides light toward a linear polarizer (GT10, ThorLabs, Newton, New Jersey) and an aperture (D20S, Thorlabs, Newton, New Jersey) to be incident on the sample. The reflected light is collected by another optical fiber ($f2$) (FT600EMT, Thorlabs, Newton, New Jersey) that guides light to a charge-coupled device spectrometer (HRS-BD1-025, Mightex Systems, Pleasanton, California). The sample is mounted on a rotatable stage that controls the incidence angle θ and the collecting end of $f2$ is mounted on a rotatable arm that moves to the angle 2θ to collect the specularly reflected light. The total transmittance of the structure is null valued in the chosen spectral regime because the metal is thicker than the penetration depth, which was experimentally verified as well. In addition, all nonspecular reflectances are also null valued for the chosen values of λ_0/L, θ, and ϕ. Therefore, the corresponding absorptances of the structure can be calculated as $A_{s,p} = 1 - R_{s0,p0}$. The measured values of the absorptances A_s and A_p are presented and discussed in Sec. 4.2.

3 Theory in Brief

3.1 *Surface-Plasmon-Polariton Wavenumbers*

The SPP wavenumbers q were obtained by solving a canonical boundary-value problem.[13,19] Suppose the half space $z > 0$ is occupied by a metal and the half space $z > 0$ by the 1-D PC. The interface can guide the propagation of an SPP wave parallel to the unit vector $\hat{\underline{u}}_{\text{prop}}$ such that $\hat{\underline{u}}_{\text{prop}} \cdot \hat{\underline{u}}_z = 0$, the associated fields varying in the xy plane as $\exp[iq\hat{\underline{u}}_{\text{prop}} \cdot (x\hat{\underline{u}}_x + y\hat{\underline{u}}_y)]$, where the Cartesian unit vectors are denoted by $\underline{u}_x$, etc. The dependences of the fields on z are in consonance with the Floquet theory, but are too complicated to be discussed here.[19]

The procedure to determine q is described in detail elsewhere.[13,19] Let us note that (1) q does not depend on the direction of $\hat{u}_{\text{prop}}$ since all materials are isotropic and the structure is transversely isotropic with respect to the z axis and (2) multiple values of q for any specific λ_0 are possible.

3.2 *Wavenumbers of Waveguide Modes*

As the 1-D PC is thicker than the free-space wavelength $\lambda_0 \in [600, 1000\ \text{nm}]$, it supports multiple waveguide modes that trap light and play a very significant light-management role for solar-harvesting applications.[16–18,24] The wavenumbers q of these modes can be obtained using a transfer–matrix approach[27] that yields the following matrix equation for a two-period PC bounded by air on one of its two faces and a perfect electric conductor (PEC) on its other face:

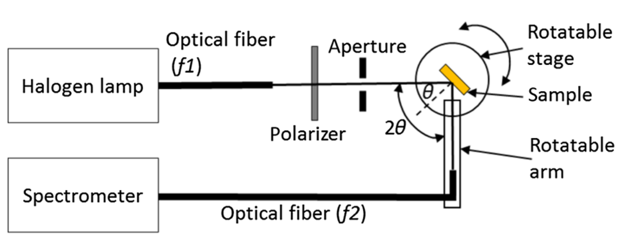

Fig. 3 Schematic of the custom spectrometer used for reflectance measurements.

$$\begin{bmatrix} 0 \\ 0 \\ 1 \\ 1 \end{bmatrix} = \{\exp(i[\underline{P}]_9 d_9) \cdot \exp(i[\underline{P}]_8 d_8) \cdot \cdots \cdot \exp(i[\underline{P}]_1 d_1)\}^2 \cdot \begin{bmatrix} \beta_p \alpha_0 / k_0 \\ \beta_s \\ \beta_s \alpha_0 / k_0 \eta_0 \\ -\beta_p / \eta_0 \end{bmatrix}. \tag{1}$$

Here, the 4×4 matrices

$$[\underline{P}]_j = \begin{bmatrix} 0 & 0 & 0 & \omega\mu_0 - q^2/\omega\epsilon_0 n_j^2 \\ 0 & 0 & -\omega\mu_0 & 0 \\ 0 & -\omega\epsilon_0 n_j^2 + q^2/\omega\mu_0 & 0 & 0 \\ \omega\epsilon_0 n_j^2 & 0 & 0 & 0 \end{bmatrix}, \quad j \in [1,9], \tag{2}$$

where $\omega = k_0 c_0$ is the angular frequency and $c_0 = 1/\sqrt{\epsilon_0 \mu_0}$ is the speed of light in free space; ϵ_0 is the permittivity, μ_0 is the permeability, and $\eta_0 = +\sqrt{\mu_0/\epsilon_0}$ is the intrinsic impedance, of free space; $\alpha_0 = +\sqrt{k_0^2 - q^2}$ is either positive real or positive imaginary; while β_p and β_s are the complex amplitudes of the p and s-polarized planewave components of light in the air side of the PC. The replacement of gold by a PEC is a reasonable simplification. Equation (1) can be cast as an eigenvalue problem to yield the wavenumbers q of p- and s-polarized waveguide modes.

3.3 *Predictions for a One-Dimensional Photonic Crystal atop a Two-Dimensional Metal Grating*

According to the Floquet theory,[28,29] when a plane wave is incident on a finitely thick 1-D PC atop a 2-D metal grating, all fields must be decomposed everywhere in terms of Floquet harmonics. For the problem under consideration here, a Floquet harmonic of order (m, n) varies with respect to x and y as $\exp[ik_0(x \cos\phi + y \sin\phi)\sin\theta] \cdot \exp[i2\pi(mx + ny)/L]$, where $m \in \{0, \pm 1, \pm 2, \dots\}$ and $n \in \{0, \pm 1, \pm 2, \dots\}$; see Chap. 3 of Ref. 19. An SPP wave or a waveguide mode with wavenumber q is excited as a Floquet harmonic of order (m, n), provided that

$$\pm\mathrm{Re}(q) \simeq k_0\{[\sin\theta + (m\cos\phi + n\sin\phi)(\lambda_0/L)]^2 + [(m\sin\phi - n\cos\phi)(\lambda_0/L)]^2\}^{\frac{1}{2}}. \tag{3}$$

4 Results and Discussion

4.1 *Theoretical Results and Predictions*

The normalized wavenumbers q/k_0 of SPP waves guided by a planar metal-PC interface are presented in Fig. 4 as functions of λ_0. These wavenumbers are organized in this figure into three branches for p-polarized SPP waves and two branches for s-polarized SPP waves.

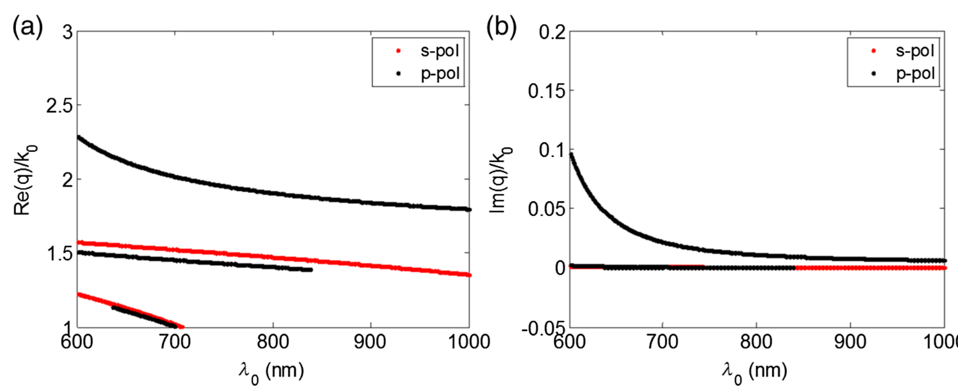

Fig. 4 (a) Real and (b) imaginary parts of the normalized wavenumbers q/k_0 of surface-plasmon-polariton (SPP) waves obtained after solving the canonical boundary-value problem.

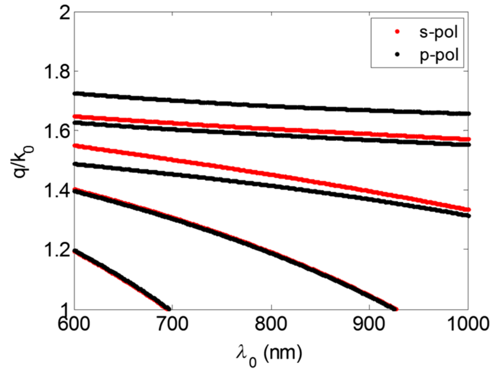

Fig. 5 The normalized wavenumbers q/k_0 of waveguide modes calculated using the transfer–matrix approach.

The normalized wavenumbers q/k_0 of the waveguide modes in the air/2-period-PC/PEC structure are presented in Fig. 5. These waveumbers are organized into five branches for p-polarized waveguide modes and four branches for s-polarized waveguide modes.

We inserted the values of q provided in Fig. 4 for SPP waves and in Fig. 5 for waveguide modes, and determined that the triplets $\{\sin\theta, m, n\}$ that would satisfy Eq. (3) for each value of q when $\phi \in \{0\text{ deg}, 45\text{ deg}\}$. Of course, a triplet can be considered as physically acceptable only if $-1 < \sin\theta < 1$. The results are plotted in Fig. 6 for $\lambda_0 \in [600, 1000\text{ nm}]$, where the branches indicate the conditions (i.e., the values of λ_0 and θ) at which the SPP and waveguide modes can be excited. It is possible for a specific SPP wave or waveguide mode to be excited as two different Floquet harmonics at the same value of λ_0; likewise, it is also possible for an SPP wave or waveguide mode of a specific polarization state to be excited by incident light of a different polarization state.[19]

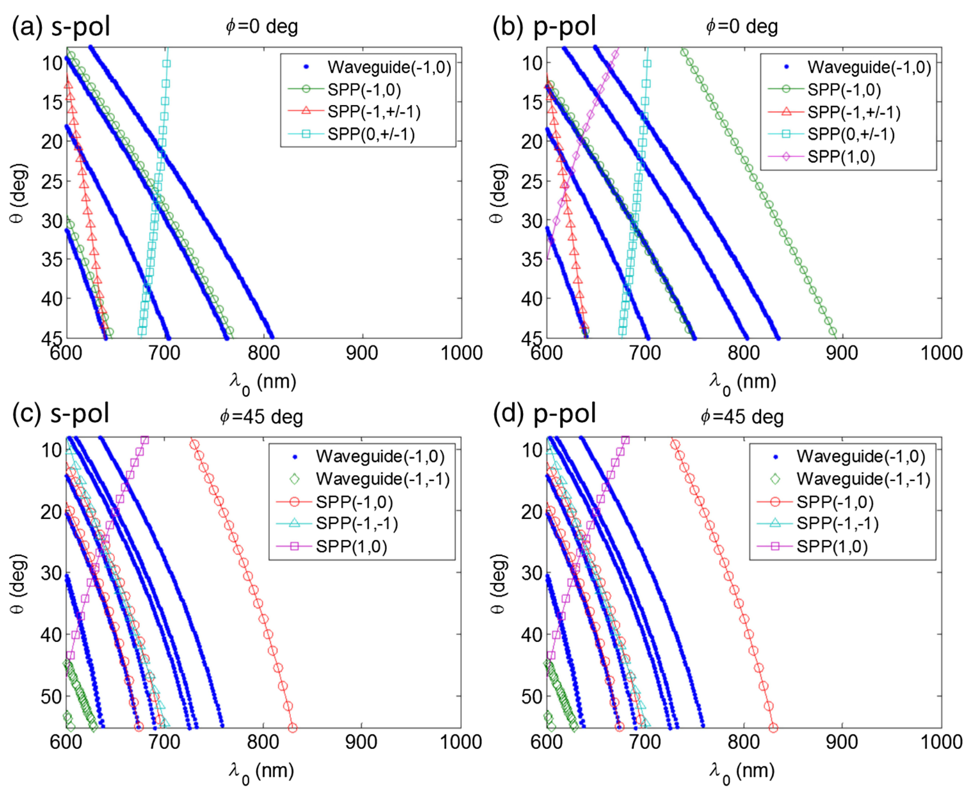

Fig. 6 Predicted values of θ obtained from Eq. (3) and Figs. 4 and 5 for the experimental excitation of SPP waves and waveguide modes of (a,c) s-polarization state and (b,d) p-polarization state for (a,b) $\phi = 0$ deg and (c,d) $\phi = 45$ deg.

4.2 Experimental Results and Correlation with Predictions

As stated in Sec. 2, we measured the specular reflectances R_{s0} and R_{p0} of the fabricated structure for $\lambda_0 \in [600, 1000\ \text{nm}]$, $\theta \in [8\ \text{deg}, 45\ \text{deg}]$, and $\phi \in \{0\ \text{deg}, 45\ \text{deg}\}$. Figure 7 presents the color maps of the absorptances A_s and A_p as functions of θ and λ_0 for the two chosen values of ϕ. Higher absorptances are shown as reddish colors, lower absorptances are shown as bluish colors.

The color maps show broadband light absorption over the chosen range of θ, and we clearly observe reddish ridges of high absorptance corresponding to the excitation of SPP waves and waveguide modes. Overall, the high-absorptance ridges overlay with the theoretically predicted curves in Fig. 6. However, one SPP wave is not observed in our experimental plots and the positions of some of the high-absorptance ridges are slightly shifted from our theoretically predicted curves. These discrepancies might be due to the distortion of the silicon-oxynitide layers in the fabricated structure. This is because these layers are conformally coated on the grating and are not necessarily perfectly planar. Another possible reason for the discrepancies is that some SPP waves are less localized than others and require that the 1-D PC be more than two periods in thickness.

On comparing our theoretical and experimental results with those for the 1-D grating,[22] we find that the 2-D grating indeed leads to the excitation of more guided waves. We observe broader-band light absorption (high absorptance for $\lambda_0 < 850$ nm) which is less sensitive to the incidence angle θ. The structure shows similar light-coupling characteristics for incident p- and s-polarized light, indicating that the absorption in the structure with a 2-D grating is less dependent on the polarization state of the incident light. A comparison of the results of $\phi = 0$ deg and $\phi = 45$ deg reveals that a rotation of the structure about the z axis will not greatly influence the absorption of light. Accordingly, the use of 2-D gratings instead of 1-D gratings as metallic back-reflectors should be more efficient for harvesting solar energy.

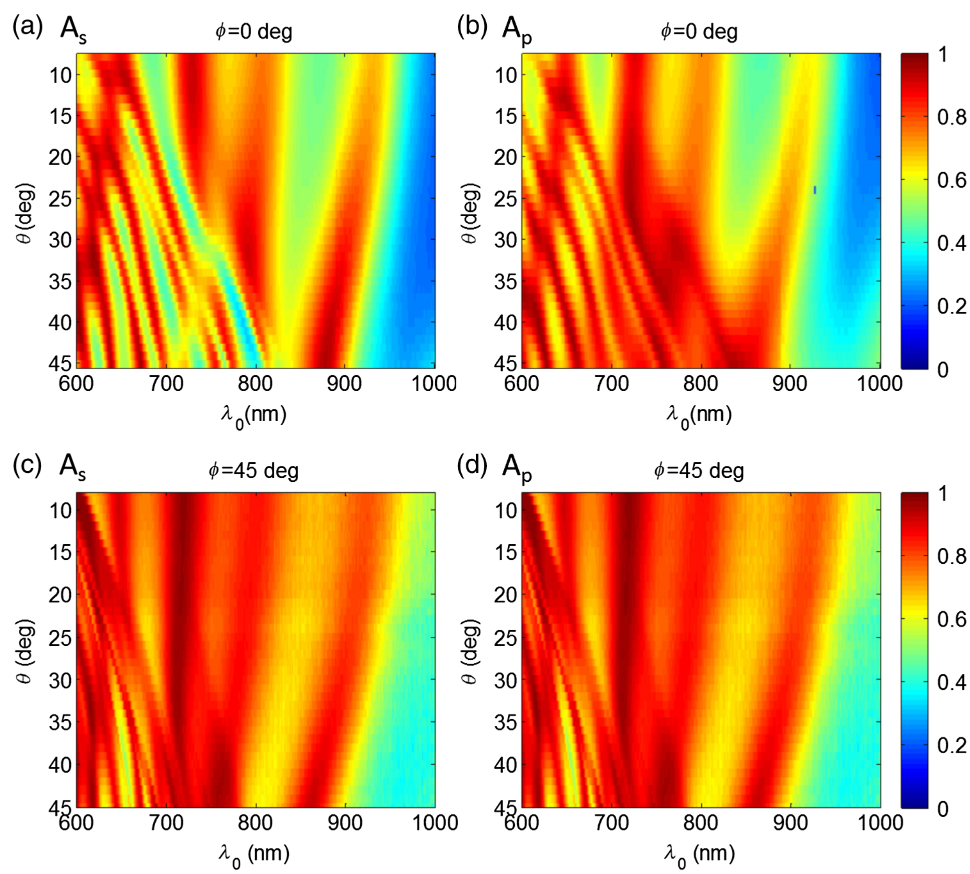

Fig. 7 Absorptances (a,c) A_s and (b,d) A_p of the fabricated structure measured as functions of λ_0 and θ for (a,b) $\phi = 0$ deg and (c,d) $\phi = 45$ deg.

5 Concluding Remarks

We deposited a 1-D photonic crystal on top of a 2-D metal grating. The absorptances of this structure for incident *p*- and *s*-polarized light were measured and mapped against the free-space wavelength and the incidence angle with respect to the thickness direction. The Floquet theory,[28,29] surface-multiplasmonics theory,[19] and the transfer–matrix approach for multilayered waveguides[27] were used to predict the excitations of SPP waves and waveguide modes as Floquet harmonics. The excitations of guided waves of both types were found to agree well with theoretical predictions.

Both the theoretical and experimental results demonstrated broadband coupling of light of any linear polarization state over a broad range of the angles of incidence. Thus, the fabricated structure can be potentially useful as a planar solar concentrator,[24] and our light-management approach will also be applicable to thin-film solar cells.[23]

Acknowledgments

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