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Azimuthal dispersion and energy mode condensation of grating-coupled surface plasmon polaritons

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The excitation conditions for surface plasmon polaritons (SPPs) on a silver-gold bilayer coated sinusoidal grating can be varied over a wide range by tuning the azimuthal grating orientation (\(\alpha\)). Grating coupling induces rotation of the SPP wave vector which, for specific conditions, can be directed perpendicular to the exciting light direction. Certain \(\alpha\) orientations allow the excitation of two SPPs with the same frequency but different propagation directions. Other azimuthal orientations allow excitation of many SPP modes characterized by propagation over a large angular range. The kinematics of SPP propagation can be described by a model based on the wave-vector conservation law. Using this model, SPP dispersion relation, propagation direction, and mode density have been computed and shown to be in agreement with experimental measurements. The wave-vector dispersion is characterized by an energy threshold for the SPP excitation that increases as \(\alpha\) increases. The angular spread is accompanied by an energy condensation of the SPP modes in correspondence to the energy threshold.

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I. INTRODUCTION

In 1902, Wood reported the observation of a series of sharp changes of reflected radiation in the spectra from ruled metallic diffraction gratings that are now known to be surface plasmon polariton modes (SPPs).1,2 SPPs are electromagnetic waves coupled with surface plasma charge oscillations that propagate along a metal-dielectric interface. These nonradiative modes have a wave vector \(k_s\) greater than the wave vector of a photon of the same frequency in vacuum. It is therefore necessary to enhance the momentum of the incident light in order to couple the SPP propagation onto the surface of the metallic film. This momentum enhancement can be provided by the presence of a diffraction grating, which modifies the in-plane incident photon wave vector by adding or subtracting multiple integers of the grating wave vector \(g\). Alternatively, several experimental setups exploit prism coupling with the well-known Kretschmann configuration.3,4

The excitation of SPP modes and their interaction with gratings have been extensively studied.5–8 Grating-coupled surface plasmon resonance (GCSPR) has also been widely used for a variety of biomedical and chemical sensing applications.9,10 Current GCSPR sensors have sensitivity on the order of \(4 \times 10^{-7}\) refractive index unit (RIU),11 almost in the sensitivity range of prism-coupled SPR.12–14 The GCSPR approach is particularly interesting for those biosensing applications that require prismless detection in an array format combining simultaneous high throughput and sensitivity. Advantages as well as drawbacks of GCSPR based techniques have been widely discussed in the literature.15

In this paper we focus our attention on a specific feature that is exclusive to grating coupling, i.e., controlling the planar propagating direction on the plane of the metallic grating. This possibility is provided by the well-known conical configuration where the incident wave vector of the incident light is not required to be parallel to the grating Bragg vector. When the grating is rotated azimuthally, symmetry is broken with an additional component due to the grating momentum. As a result, the final SPP propagating direction is no longer perpendicular to the groove but rotated instead. Breaking the collinear symmetry greatly enlarges the set of physical combinations that satisfy SPP coupling. In particular it has been demonstrated that a fraction of \(p\)-polarized light is converted to \(s\)-polarized light,16 an effect that has been fully explained using computational,17,18 and recently, fully analytical approaches.19 Studies on conical mounting include characterization of metal gratings,20 narrow-ridged short-pitch metal gratings in the conical mount,21 sharp resonances,22 waveguiding,23 and out coupling of SPPs to radiation modes in the grating-prism combined configuration.24

However, notwithstanding the high interest for GCSPR, little attention has been paid to the direction and the number of different modes that can be excited with the conical configuration. In this paper we specifically investigate the conditions for SPP excitation as a function of azimuthal rotation angle on the grating plane. We show a new way to use a single incident wavelength to excite two SPPs with different propagation directions. Particularly interesting are the conditions for the merging of the two SPPs that propagate perpendicularly to the incoming light. The wave-vector conservation provides a simple description of the kinematics of SPP grating coupling and also allows determination of the wave-vector direction of rotation and the related energy dispersion. Furthermore the angular spreading of SPP wave-vector directions can be described and related to the number of SPP
modes that can be excited by a single wavelength. Azimuthal orientation allows the possibility to simultaneously excite a large number of SPP modes with the same energy but different orientations. This effect has been related to an energy density condensation mode that occurs in correspondence to the energy threshold for SPP excitation. The implications of these findings in the context of sensing applications will be described.

II. EXPERIMENT

The gratings used in this study were produced by the following technique: Flat PMMA slabs have been imprinted using sinusoidal grating generated by holographic lithography methods described elsewhere.25,26 Due to the high coherence length of holographic lithography patterning, highly uniform periodic gratings can be obtained over a large region (20 × 20 mm²). A double layer film of Ag and Au has been evaporated on top of these gratings with a thickness of 37 and 7 nm in succession. The grating used in this study has an almost sinusoidal profile with periodicity of 476 nm and amplitude of 29 nm.

The optical measurements have been performed in θ/2θ symmetric reflectivity configuration, scanning angular position with a precision of 0.005° and using a 75 W Xe lamp, monochromatized between 600 and 900 nm with a bandwidth of 2.7 nm. All measurements were collected at p polarization. In our experimental setup, the scattering plane is fixed and the samples with gratings were mounted on a sample holder that can be rotated azimuthally to define the α angle. The grating’s orientation was varied by rotating azimuthally from 0° (i.e., grating perpendicular to incident light) to 60° with a precision of 0.01°. All measurements were acquired using a J.A. Woollam Co. VASE instrument.

III. RESULTS

The first evidence of the azimuthal rotation effect on the SPR excitation is shown by the reflectivity spectra collected at different azimuthal grating rotations (Fig. 1). The azimuthal rotation is accompanied by clear shift of the resonance angle position and by an increase in the width of resonance. However, this effect changes dramatically when azimuthal rotation exceeds a critical value, which in our case is αc=45.62°. The SPR reflectivity spectra in Fig. 2 were collected at α=52.5° with light of different wavelengths ranging from 602 to 625 nm. It is clearly shown that two minima can be excited using wavelengths up to 610 nm. The two minima approach each other as the excitation wavelength increases, and finally merge into a single broad dip at 612 nm with a full width at half minimum (FWHM) of ΔθFWHM=28°. Further increment in the wavelength results in shallower reflectivity dips, and the dips finally disappear at wavelengths larger than 625 nm.

The described phenomena are confirmed by the full set of SPR data (λ, θmin) in Fig. 3. We have also reported the FWHM reflectivity dip width, ΔθFWHM, whose amplitude increases with the azimuthal angle. The guiding lines clearly show that by increasing the azimuthal rotation, the slope of wavelength vs incidence angle decreases. For azimuthal angles greater than a critical value αc, a single wavelength can excite two SPPs. Further increase in wavelength will merge the two SPPs into a single broad dip until a maximum wavelength above which SPP excitation is prevented.

IV. DATA ANALYSIS

The mechanism of polariton excitation in grating-coupled SPR can be understood by considering that the periodic grating changes the wave vector of the incident radiation by adding an integer multiple of the grating wave vector $\vec{g}$. The
the SPP dispersion relation becomes

\[ \tilde{k}_{sp} = \tilde{k}_{sp} + m\tilde{g}, \quad (1) \]

where \( \tilde{k}_{sp} \) is the diffracted SPP wave vector and \( m \) identifies the diffraction order. The scattering plane \((x,y)\) is normal to the grating plane \((x,z)\) and contains wave vector of the incident light direction that forms an angle \( \theta \) with the normal \( z \). The light wave vector is represented by \( \tilde{k}_{sp} = 2\pi/\lambda(-\sin \theta, 0, \cos \theta) \). In our frame of reference, the grating momentum \( \tilde{g} = 2\pi/\Lambda(\cos \alpha, \sin \alpha, 0) \), which lies in the \((k_x, k_y)\) plane, is inversely proportional to the grating pitch \( \Lambda \), and is oriented perpendicular to the grooves of the gratings whose rotation is measured by the azimuthal angle \( \alpha \). The direction of the diffracted SPP wave vector \( \tilde{k}_{sp} \) is in the grating plane and is identified by the angle \( \beta \) [see Fig. 4(a)]. The SPP wave-vector magnitude is related to its wave frequency by the dispersion relation

\[ k_{sp}(\omega) = \frac{\omega}{c} \sqrt{\frac{\varepsilon_1 \varepsilon_2}{\varepsilon_1 + \varepsilon_2}}, \quad (2) \]

where \( \varepsilon_1 \) and \( \varepsilon_2 \) are the dielectric constants, respectively, of the dielectric and of the metal. In the case of light traveling in vacuum \((\varepsilon_1=1)\) and incident on a metallic medium with plasma frequency, \( \omega_p \), and dielectric constant \( \varepsilon_2 = 1 - \omega_p^2/\omega^2 \), the SPP dispersion relation becomes

\[ k_{sp}(\omega) = \frac{\omega}{c} \sqrt{\frac{\omega^2 - \omega_p^2}{2\omega^2 - \omega_p^2}}, \quad (3) \]

The \( x \) components of \( \tilde{k}_{sp} \) are given by the relation

\[ k_{sp_{x}} = k_{sp}(\omega) \cos \beta = \pm \sqrt{k_{sp}(\omega)^2 - (mg \sin \alpha)^2}, \quad (4) \]

where we used the conservation of wave vector along the \( y \) component, i.e.,

\[ k_{sp_{y}} = mg \sin \alpha. \quad (5) \]

In Fig. 4(a) the large circle in \( k \) space represents equimagnitude \( \tilde{g} \) vectors at different azimuthal orientation. The dashed line represents the \( x \) component of the incident light wave vector. The smaller circle represents all possible \( \tilde{k}_{sp} \) vectors with equal magnitudes. This is indeed the case in our experiment because the grating momentum \( \tilde{g} \) is always greater than the incident light momentum in the wavelength regime that we used for our experiments. A further consequence of our experimental setup is that only the first diffraction order is relevant \((m=1)\).

Equation (1) allows the transformation of the experimental \((\lambda, \theta_{\text{min}})\) sets of reflectivity minima into a set of points of an energy vs wave vector plot. In particular, the experimental values of the \( x \) component of SPP have been obtained using the following relation:

\[ k_{sp_{x}} = g \cos \alpha - \frac{2\pi}{\lambda} \sin \theta. \quad (6) \]

The experimental data of the SPP excitation grouped for different \( \alpha \) angles have been compared with the relations provided by Eq. (4). The experimental data and model dispersion curves have been plotted in Fig. 5(a).

For each set of data points a single azimuthal \( \alpha \) angle has been used as a fitting parameter. The value of the pitch of the grating was initially determined from AFM and used as a constant in Eq. (4). Afterwards, we keep the \( \alpha \) angles values fixed, testing the convergence on the lattice parameter finding a fitting value of \( \Lambda = 475 \pm 2 \) nm, compatible with the initial AFM determination. The independent fitting of both lattice parameter and azimuthal angle for each set of data converged at the same values previously obtained and the resulting values have been reported both in Figs. 3 and 5(a). The good agreement of the model with the experimental data provides a confidence level on the overall data analysis method.

The angle defining the \( \tilde{k}_{sp} \) direction can be determined by

\[ \beta = \tan^{-1} \left( \frac{k_{sp_{y}}}{k_{sp_{x}}} \right). \quad (7) \]

The experimental values of the SPP rotation can be obtained substituting in Eq. (7) the Cartesian component of \( \tilde{k}_{sp} \) provided by Eqs. (5) and (6). Likewise, substitution of Eqs. (4) and (5) in Eq. (7) yields the modeled analytical dependence. The energy dispersion of experimental data and the model SPP direction have been reported in Fig. 5(b). The \( \alpha \) angles used for the SPP direction are the same ones used for the determination of \( k_{sp_{x}} \) reinforcing the confidence of the fitting method.

The error bars in Fig. 3 represent the FWHM angular width, \( \Delta \theta_{\text{FWHM}} \), measured from the SPR reflectivity dips. Using Eqs. (4) and (5), \( \Delta \theta_{\text{FWHM}} \) has been transformed into
the spread of wave vectors $\Delta k_{sp}$ and of the SPP angle direction $\Delta \beta$, and have been plotted as error bars, respectively, in Figs. 5(a) and 5(b).

It is clear that $\Delta \theta_{\text{FWHM}}$ is increasing with azimuthal angle rotation and this behavior is reflected also in $\Delta k_{sp}$ and $\Delta \beta$. It is worth noting that the agreement between the model curves and the experimental data is much better than the error bar spread. As a consequence, the SPR reflectivity width is providing meaningful physical information about the number of SPP modes that can be excited. The quantity that measures the number of SPP modes (per unit of illuminated area) that are excited in a specific experimental condition is given by the differential area in the $k$ space centered on $k_{sp}$, which can be computed in polar coordinates as

$$\delta k_{sp}^2 = |k_{sp}\delta k_{sp}\delta \beta|.$$  

(8)

It is worthwhile to normalize this to the differential area of a circle of radius $k_{sp}$:

$$\delta f(\pi k_{sp}^2) = \frac{|\delta \beta|}{2\pi},$$

(9)
i.e., area covered in the $k$ space by the circular band of $|\delta k_{sp}|$ width around the circumference of $|k_{sp}|$ radius. The normalized fraction of polariton modes that can be excited together with mode $k_{sp}$ is therefore given by the ratio of the Eqs. (8) and (9):

$$f(k_{sp}) = \frac{\delta k_{sp}^2}{\delta f(\pi k_{sp}^2)} = \frac{|\delta \beta|}{2\pi}. $$

(10)

Equation (10) states that the fraction $f$ of excited SPP modes is proportional to the error bar reported in Fig. 5(b). In other words, the spread of SPP azimuthal direction gives an indication of how many modes that can be excited in a specific configuration.

To complete the computation, we substitute Eq. (7) in Eq. (10) and after differentiating we obtain

$$f = \frac{1}{2\pi} \frac{|k_{sp}\delta k_{sp}|}{k_{sp}^2} - \frac{|k_{sp}|}{k_{sp}^2} \delta k_{sp} \delta \beta.$$ 

(11)

The terms in Eq. (11) are given by:

$$(k_{sp}, \delta k_{sp})^2 = \sum_{\eta} \left( \frac{\partial k_{sp}(\eta)}{\partial \eta} \right)^2 \delta \eta.$$ 

(12)

where $i, j = x, y$, whereas $\eta$ represents the set of independent variables of $k_{sp}$ function. $f$ and its model analytical energy dependence has been computed substituting, respectively, Eqs. (5) and (6) in Eq. (12). The full set of data for $\theta_{\text{min}}, \Delta \theta_{\text{FWHM}}$ and the experimental values of $\delta \eta$, namely, $\delta \alpha = 2.7$ nm, $\delta g / g = 5 \times 10^{-4}$, and $\delta \beta = 0.015^\circ$, have been used for the determination of the experimental values of $f$ and the results for the different set of azimuthal angles are reported in Fig. 5(c). In order to determine the analytical behavior of $f$, a similar procedure has been applied to Eq. (4), using the same values of $\delta g / g$ and $\delta \alpha$ and the natural spread of the SPP $|\delta k_{sp}| = 0.0015$ nm. This latter value has been obtained experimentally, using the $\Delta \theta_{\text{FWHM}}$ value for $\alpha = 0^\circ$, where the SPP spreading is not affected by azimuthal effects. The analytical curves have also been reported for
FIG. 5. (Color online) (a) Dispersion curve of the x component of the SPP wave vector. (b) Dispersion curve of the azimuthal SPP direction. (c) Normalized plasmon mode density where the energy SPP thresholds are shown. Similar colors in all plots represent different azimuthal orientations $\alpha$. Error bar in plots (a) and (b) represent wave vector and angular SPP spread, respectively. Experimental (points) and model curves (solid lines) are compared.

comparison in Fig. 5(c). For ease of comparison, the dispersion curve of SPP wave vector, SPP direction, and SPP normalized mode density are reported in Fig. 5 on a common energy axis.

V. DISCUSSION

We use the pictures shown in Fig. 4 to explain the double SPP excitation. The scheme of Fig. 4(a) represents the projection on the $(k_x, k_y)$ plane of the dispersion curve of SPP [Eq. (4)] that is shown in Figs. 4(b) and 4(c), where it is represented in a three-dimensional plot $(k_x, k_y, E)$. Because of the plus and minus conditions inherent in the wave-vector conservation requirement [Eq. (1)], all quadrants of the circle can be explored for plasmon resonances as long as the conservation is satisfied. However for symmetry reason, only the $k_x$ positive half space is shown.

In Fig. 4(b) the large circle in $k$ space represents equimagnitude $g$ vectors at different azimuthal orientation as well as in Fig. 4(a). The origin of $k_{ph}$ is positioned at the tip of the circle of radius $g$ to take into account the crystal momentum contribution. The oblique and the vertical lines represent, respectively, the light dispersion curve in the case of surface parallel ($\theta=90^\circ$) and perpendicular ($\theta=0^\circ$) direction. The horizontal line represents the range of the $x$ component of the incident light $k_{ph}$ for a selected energy (in the picture arbitrarily chosen at 2 eV). This line is scaled linearly in $\sin \theta$ such that the full length of the line at the incident angle $\theta_{in}$ of 90° corresponds to the maximum value of $k_{ph}$. The projection of this line on the $(k_x, k_y)$ plane is also reported. In this picture, point B shows the case for $\alpha=18.7^\circ$ (one of the experimental conditions). A successful SPP excitation corresponds to the condition for which the wave-vector conservation [Eq. (1)] is satisfied; that is graphically represented by the point of intersection between the horizontal line $k_{ph}$ and the three-dimensional (3D) plasmon dispersion curve. This intersection also allows identification of the energy and the wave vector of the SPP as well as the plasmon propagation direction $\beta$. For example at point B, the photon wave vector can intersect the SPP circle only at $B_+^+$, thus exciting only a single SPP. This is also true for $\alpha=0^\circ$ [point A in Fig. 4(a)], for the common nonconical configuration, where the grating, photon, and SPP wave vectors are all parallel.

The condition of double SPP excitation is shown in Fig. 4(c) for $\alpha=55.7^\circ$ (the largest $\alpha$ value in the experiment). In this case, starting from point C on the $g$ circle, the $k_{ph}$ can intersect the SPP dispersion curve twice providing the excitation of two SPPs at two possible conditions: $C_{sp}^+$ and $C_{sp}^-$ with the propagation directions of $\beta_{sp}^+$ and $\beta_{sp}^-$, respectively. It is remarkable that the symmetry properties clearly show that propagation angles satisfy the relationship, $\beta^+ + \beta^- = 180^\circ$.

Within the double SPP range (points $C_{sp}^+$ and $C_{sp}^-$), a small increment in $\alpha$ at the same wavelength or small increment in wavelength at same $\alpha$ will cause the two $k_{sp}^+$ to merge, corresponding to the situation where two reflectivity dips merge into a single broad dip, as reported in Fig. 2. This “SPP merging” is described by the condition $\beta^+ = \beta^- = 90^\circ$, meaning that the resulting SPP propagate along the y direction which is perpendicular to the incident light.

There is another interpretation of the SPP merging that takes into account, instead of the excitation of the SPP, the exciting light condition. The SPP merging is equivalently described by the condition:

$$\tan \alpha = \frac{m g}{k_{ph}} \sin \alpha. \quad (13)$$

This represents the geometrical condition in which $k_{ph}$ is tangential to the SPP dispersion curve [point D in Fig. 4(a)]. In this condition the incident light wave vector can explore tangentially the natural width of the SPP circle $\delta k_{sp}$ and therefore intersect a larger area. This explains the larger azimuthal spreading and number of excited SPP modes.

The smallest angle for which SPP merging can be achieved corresponds to the critical angle $\alpha_c$ that is given in Eq. (13) used the largest value of $k_{ph}$, namely, when incident light is ideally parallel to the grating surface at $\theta=90^\circ$. That
depends on the magnitude of crystal vector; in our case the value of $\alpha_c = 45.62^\circ$, as previously reported. Further increasing of the $\alpha$ rotation or shrinking of $k_y$, circle makes the $y$ component of the $\vec{g}$ vector larger than the SPP wave vector $mg \sin \alpha > k_y$, and thus no intersection can occur (point $E$). This corresponds to a condition where plasmon excitation is not possible.

The $x$ component of the SPP curves for different azimuthal rotation angles is shown in Fig. 5(a). The curves represent sections of the 3D SPP dispersion curve obtained at different values of $k_z = g \sin \alpha$, as shown in example in Figs. 4(b) and 4(c). The higher the $\alpha$ angle, the higher and smoother it appears in the SPP dispersion curve. For $\alpha > \alpha_c$, it is possible to achieve the SPP merging at the minimum of the SPP dispersion curve. This fact is better shown in Fig. 5(b), reporting the dispersion curve of the SPP azimuthal angle where the energy minima occurs at $\beta = 90^\circ$. For each $\alpha$ value, the corresponding SPP direction varies over a large range of $\beta$ angle. In particular, for the largest azimuthal rotation used in our experiment ($\alpha = 55.7^\circ$) we measured SPP azimuthal rotation well over $90^\circ$, reaching a value up to $\beta = 105^\circ$.

The error bar reported in Fig. 5(a) shows that the SPP mode spread increases when the $\alpha$ angle is increased. This effect has a simple physical explanation that is based on Eq. (10) and by the interpretation of the corresponding error bar in Fig. 4(b). The number of SPP modes excited is proportional to their azimuthal direction distribution. In Fig. 5(c) we have reported as a function of energy both the experimental (points) and analytical (lines) values of the normalized SPP mode density $f$, clearly showing a reasonable agreement. The analytical description shows a singularity corresponding to the minimum energy for double SPP excitation, where the agreement with experiment is expectedly less.

As is well-known, a higher number of modes is expected for a flat energy band. The number of SPP modes is proportional to the inverse of the band curvature at the dispersion curve energy minima. Experimentally, this is clearly shown as error bars becoming larger for flatter energy dispersion curves, with a correspondingly increasing number of modes. For fixed $\alpha$ value, the larger value of the error bar has been determined in correspondence of the energy minimum, namely, in correspondence of the SPP merging condition. The largest error bar is achieved at SPP minimum of the largest $\alpha$ value. These results clearly show that the possibility to excite several SPPs over a broad incidence angle is accompanied by a mode density condensation in correspondence of the energy threshold.

VI. CONCLUSIONS

In summary, we have studied the kinematics of SPP propagation as a function of the azimuthal rotation $\alpha$ of the metallic grating. Azimuthal rotation of grating also affects the dispersion curves of the SPP wave vector in a way which is fundamentally different from that of classical prism coupling. The grating azimuthal rotation induces rotations of the SPP propagation direction that can cover the full $2\pi$ range. In particular conditions of incident wavelength and grating azimuthal orientation, distinct double SPPs can be excited and even multiple SPPs can propagate. The wave-vector dispersion is characterized by an energy threshold for the SPP excitation that increases as $\alpha$ increases. For azimuthal rotation larger than a critical value, the SPP energy threshold corresponds to the excitation of SPPs directed perpendicularly to the incident light. Moreover, they correspond to a divergence of the wave-vector derivative and therefore to a maximum of the SPP density of states. We have shown that the possibility to excite several SPPs over a broad azimuthal angle is accompanied by a mode density condensation in correspondence of the energy threshold. We believe that this optimized coupling condition can be used as amplification leverage for enhancing sensitivity in GCSPR systems.

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