Interference of conically scattered light in surface plasmon resonance

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Surface plasmon polaritons on thin metal films are a well studied phenomena when excited using prism coupled geometries such as the Kretschmann attenuated total reflection configuration. Here we describe a novel interference pattern in the conically scattered light emanating from such a configuration when illuminated by a focused beam. We observe conditions indicating only self-interference of scattered surface plasmon polaritons without any contributions from specular reflection. The spatial evolution of this field is described in the context of Fourier optics and has applications in highly sensitive surface plasmon based biosensing. © 2013 Optical Society of America

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In the Kretschmann attenuated total reflection (ATR) configuration, excitation of surface plasmon polaritions (SPPs) is known to produce a minimum [1] in the angular intensity of reflected light about the surface plasmon resonance (SPR) condition. This minimum is attributed [2] to an interference effect between a specularly reflected component and an antiphase reradiated plasmon field. For a focused beam incident at the SPR angle with its (tilted) diffraction limited spot at the interface, the reflected field contains a spectrum of **k**-vectors resulting in an angular intensity spectrum that can be observed in the far-field.

In this situation it is interesting to note that, because surface plasmons are excited at the focus, the system can be seen in the context of Fourier optics as a spatial filter which modifies its local **k**-vectors. The reflected field is then essentially a spatial manifestation of the surface plasmon's resonance. This Fourier optics picture of SPR turns out to be remarkably intuitive for describing this system's behavior. In the far-field (Fraunhoffer) regime, the field on the interface and the far-field pattern are conjugate variables related by a simple Fourier transform pair. In the Fresnel regime, a sharp resonance will act as a low-pass filter for light, resulting in curious one-sided spatial oscillations in the reflected signal, which evolve with propagation into the far-field.

Somewhat surprisingly perhaps, despite the ubiquity of this configuration as a platform for biosensing, one-sided oscillations in the Fresnel regime were not predicted until as recent as 2005 [3,4], and to the authors' knowledge have only been experimentally reported in two limited cases [5,6]. In the cited literature, these oscillations have been described as being phenomenologically similar to the SPR intensity minimum in the far-field: an interference effect between a specularly reflected (ϵ_1-ϵ_2 interface) beam and a reradiated plasmon field (ϵ_2-ϵ_3 interface).

In this Letter we present evidence that such behavior is more aptly described as a general diffraction phenomena and *does not require* two interfering components. To this accord we present the first observation of spatial oscillations in the conically scattered light from SPPs on thin metal films in the Kretschmann ATR configuration.

The experimental geometry is shown in Fig. 1. It consists of a 48 nm silver film sputtered on to the hypotenuse of a LAH79 hemispherical prism. A p-polarized Gaussian beam with a $1/e^2$ beam waist of $w_0 = 3$ mm from a 632.8 nm helium–neon laser is directed through a $f_{1,2} = 25$ mm lens and focused to the central point of the hypotenuse of the prism at the SPR angle $\theta_{\mathrm{SP}} \approx 32°$. The totally internally reflected or scattered light from different focal planes is then imaged by f_2 onto an image sensor, which records the resulting optical profile.

Upon interaction with the prism, the incident light is directed into two directions. The first is the specular direction, which is the most common way to observe SPR and is the direction where the majority of light is present. This direction includes a specularly reflected component from the ϵ_1-ϵ_2 interface and a reradiated plasmon field from the ϵ_2-ϵ_3 interface. Since the light from the ϵ_1-ϵ_2 interface is antiphase with that from the ϵ_2-ϵ_3 interface

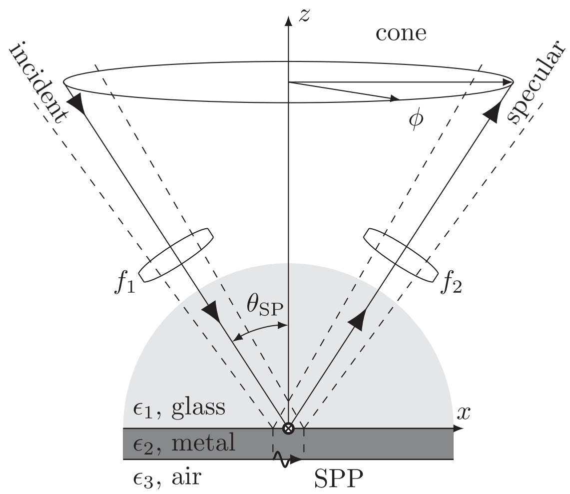

Fig. 1. Experimental setup. Light is incident from the left and focused by a lens f_1 on to the hypotenuse of a hemispherical prism coated with a thin layer of metal (Ag). The majority of light is directed into the specular direction. Surface roughness scatters SPPs on the ϵ_2-ϵ_3 interface, causing reradiation of the plasmon field into a hollow cone. f_2 acts to image light exiting the system at different focal planes.

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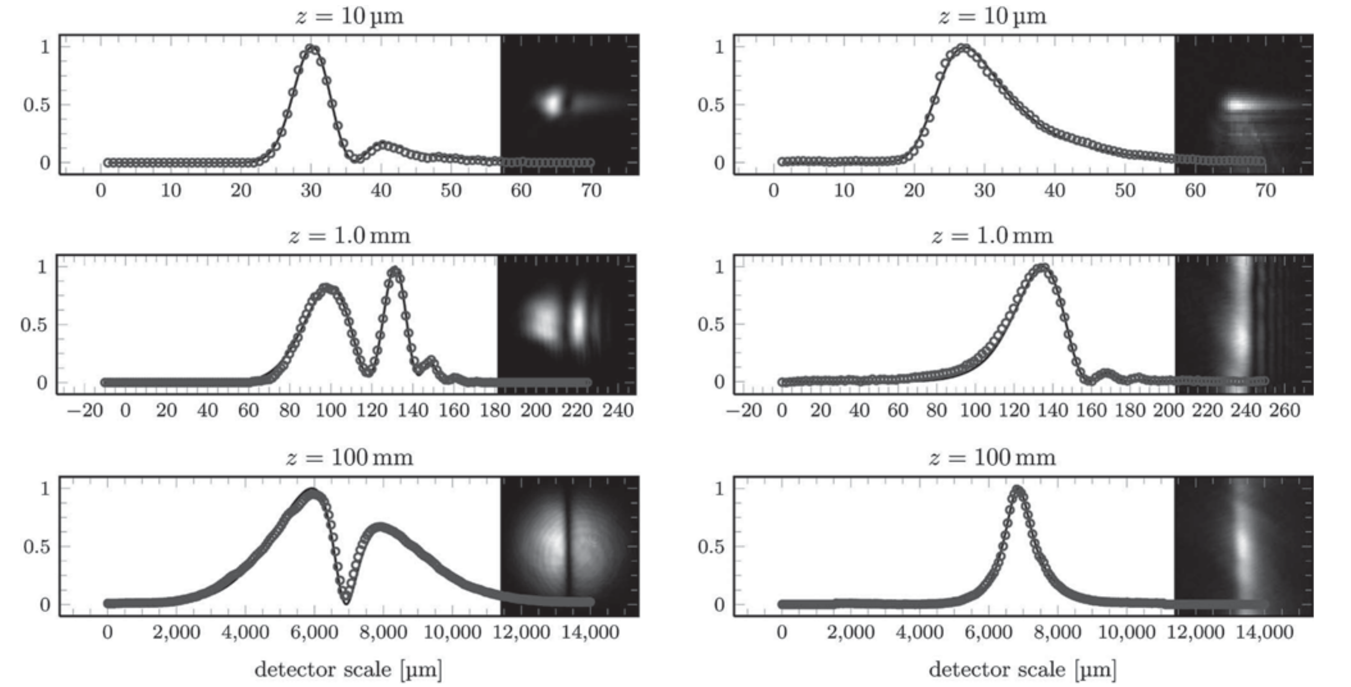

Fig. 2. Theoretical and experimental values of $|E_{\mathrm{spec}}(x,z)|^2$ and $|E_{\mathrm{cone}}(x',z)|^2$ obtained for $z = 10$ μm, $z = 1.0$ mm, and $z = 100$ mm. Theoretical values are solid curves, while experimental values are shown with circles. Each plot has been normalized independently for comparison. The normalized two dimensional output from the image sensor is inset in each plot.

at θ_{SP}, the resulting spatial profile takes on a notched Gaussian appearance (see measured intensity profile in Fig. 2, left column, $z = 100$ mm). The second direction concerns scattered light. When an SPP is excited on the ϵ_2-ϵ_3 interface, surface roughness can elastically modify the SPP's in-plane momentum (k_x or k_y). Since both the magnitude and direction of this momentum must be conserved, light from this field falls into a hollow cone.

We first consider light directed to the specular direction. This has been previously reported by [3,4], and we shall use a similar treatment. Restricting the analysis to the x–z plane, the angular profile can be described using the Fresnel reflectivity for a three layer system [7], $\tilde{r}_{123}(k_x)$ (ϵ_1-ϵ_2-ϵ_3), multiplied by a Fourier decomposed incident Gaussian beam $\tilde{g}(k_x)$

$$\tilde{E}_{\mathrm{spec}}(k_x) = \tilde{g}(k_x)\tilde{r}_{123}(k_x). \tag{1}$$

The complete spatial profile in both x and z can be obtained by computing the Fourier transform of $\tilde{E}_{\mathrm{spec}}(k_x)$ multiplied by a free space transfer function $\mathrm{e}^{\mathrm{i}k_z z}$:

$$E_{\mathrm{spec}}(x,z) = \int_{-\infty}^{\infty} \tilde{E}_{\mathrm{spec}}(k_x)\, \mathrm{e}^{\mathrm{i}k_z z}\mathrm{e}^{\mathrm{i}k_x x}\mathrm{d}k_x, \tag{2}$$

where $k_x = k_0\sqrt{\epsilon_1}\,\sin\theta$ and $k_z = \sqrt{k_0^2\epsilon_1 - k_x^2}$. Equation (2) is shown as a function of x and z in Fig. 2. At $z = 10$ μm, $|E(x, z = 10\ \mu\mathrm{m})|^2$ qualitatively matches the surface field theoretically predicted via a similar method by [7], or through vector Gaussian beam decomposition [8]. Our measurements of the intensity profile in the intermediate and far-field regimes (Fig. 2, left column, $z = 1.0$ mm and $z = 100$ mm) also agree very well with predictions based on Eq. (2).

As mentioned, the origin of the one-sided oscillations has previously been described as arising due to the interference between the specular reflection from the ϵ_1-ϵ_2 interface and reradiated plasmon field from the ϵ_2-ϵ_3 interface. While it is true that the field in the specular direction contains both components, and that $\tilde{r}_{12}(k_x)$ (ϵ_1-ϵ_2) and $\tilde{r}_{23}(k_x)$ (ϵ_2-ϵ_3) are indeed antiphase at θ_{SP} causing interference, the one-sided oscillatory pattern does not require a specular component to be observed, as we will show by looking at optical patterns observed for light scattered into the cone.

Consider scattered light at a position (x', z) within the scattering cone, $E_{\mathrm{cone}}(x', z)$, where x' is the original x domain rotated azimuthally by ϕ (Fig. 1). Experimentally this may be observed at any location that is not coincident with the incident or specularly reflected beams. The angular intensity distribution in ϕ is nearly isotropic and contains speckle consistent with a tightly focused beam and a slightly rough metal film [5]. The intensity of light scattered into the cone is approximately 0.1% that of the incident beam [9].

Its intensity profile is given by the Fresnel transmittance $\tilde{t}_{123}(k_{x'})$ of the Gaussian beam $\tilde{g}(k_{x'})$, representing light passing through the system and exciting SPP waves on the ϵ_2-ϵ_3 interface, multiplied by $\tilde{t}_{321}(k_{x'})$, representing the reverse path through the system:

$$\tilde{E}_{\mathrm{cone}}(k_{x'}) = \tilde{g}(k_{x'})\tilde{t}_{123}(k_{x'})\tilde{t}_{321}(k_{x'}), \tag{3}$$

$$E_{\mathrm{cone}}(x', z) = \int_{-\infty}^{\infty} \tilde{E}_{\mathrm{cone}}(k_{x'})\mathrm{e}^{\mathrm{i}k_z z}\mathrm{e}^{\mathrm{i}k_{x'}x'}\mathrm{d}k_{x'}. \tag{4}$$

The predicted evolution in the propagation of $|E_{\mathrm{cone}}(x', z)|^2$ along with its experimentally observed counterpart is shown for three selected distances in Fig. 2, right column. Like $|E_{\mathrm{spec}}(x, z)|^2$, $|E_{\mathrm{cone}}(x', z)|^2$ too exhibits one-sided oscillations, but in this case it is lacking any component of light specularly reflected from the ϵ_1-ϵ_2 interface. In the far-field, this profile resembles an inverse of the notch in the specular direction. Since the light scattered into the cone lacks a specular component, its origin can be seen as arising from a situation akin to the classic Fresnel edge diffraction. Here the presence of a rapid change in the system's transmission/reflection (an "edge"), acts as a low pass filter for light, and the resulting truncated Fourier integral leads to spatial oscillations due to Gibb's phenomena. In this respect the behavior is clear if one considers that the Fresnel relations used to model the system are casual functions. As proof, take a complex function $\chi(\omega) = \chi'(\omega) + \mathrm{i}\chi''(\omega)$ whose real and imaginary parts are related by Kramers–Kronig relations,

$$\chi(\omega) = \mathrm{i}\mathscr{H}^{+}\Big(\chi(\omega)\Big), \tag{5}$$

with

$$\chi'(\omega) = \mathscr{H}^{+}\Big(\chi''(\omega)\Big), \tag{6}$$

$$\chi''(\omega) = -\mathscr{H}^{+}\Big(\chi'(\omega)\Big), \tag{7}$$

where $\mathscr{H}^{+}(\chi(\omega))$ is the Hilbert transform of $\chi(\omega)$. The Fourier transform of $\chi(\omega)$ is

$$\mathcal{F}^{+}\chi(\omega) = \mathcal{F}^{+}\Big(\chi'(\omega) + \mathrm{i}\chi''(\omega)\Big), \tag{8}$$

$$= \mathcal{F}^{+}\Big(\chi'(\omega)\Big) + \mathrm{sgn}(\omega)\mathcal{F}^{+}\Big(\chi'(\omega)\Big). \tag{9}$$

Or succinctly,

$$\mathcal{F}^{+}\Big(\mathscr{H}^{+}\Big(\chi(\omega)\Big)\Big) = \Big(-\mathrm{i}\,\mathrm{sgn}(\omega)\Big)\mathcal{F}^{+}\Big(\chi(\omega)\Big). \tag{10}$$

In other words, the Fourier transform of any function, which satisfies Kramers–Kronig relations is "one-sided" as a necessary condition of causality.

Theoretical analysis [10] of the ultimate resolution of SPR for bulk index sensing seems to indicate that it is agnostic with regards to both the coupling principle (prism, grating) and interrogation method (angular, intensity, wavelength, or phase). This analysis was based on the far-field pattern in the specular direction. It is an interesting question whether the same analysis applies in the presence of spatial oscillations, and which, if any such properties may lend themselves to advancing the resolution of SPP based biosensors. In passing, we note that for small refractive index perturbations, the shift of the spectral features is approximately linear, and if referenced to the width of a propagated Gaussian beam, the spatial oscillations represent slightly sharper angular features than the single far-field resonance in classic SPR.

In conclusion, we have applied Fourier optics principles to the specularly reflected and conically scattered light for SPP excitation on thin metal films in the Kretschmann ATR configuration. In doing so we have predicted and observed a new interference phenomena in the conically scattered light caused by the causal response of reradiated SPPs. All observations are in excellent agreement with theoretical predictions. These spatial oscillations may provide extra features to increase the fidelity of current SPR based measurements.

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