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Leakage radiation microscopy of surface plasmon polaritons

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Abstract

We review the principle and methodology of leakage radiation microscopy (LRM) applied to surface plasmon polaritons (SPPs). Therefore we first analyze in detail the electromagnetic theory of leaky SPP waves. We show that LRM is a versatile optical far-field method allowing direct quantitative imaging and analysis of SPP propagation on thin metal films. We illustrate the LRM potentiality by analyzing the propagation of SPP waves interacting with several two-dimensional plasmonic devices realized and studied in the recent years.

Keywords: Surface plasmons; Polaritons; Lekage radiation microscopy; Optical far field; Imaging

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1. Introduction

In recent years intensive investigations of surface plasmon polaritons (SPPs) have been made in the promising context of nanophotonics. This research is actually motivated by the current trends for optical device miniaturisation and by the possibilities of merging aspects of nanophotonics with those of electronics. SPPs are electromagnetic waves bounded to dielectric–metal interface. As surface waves, SPPs are exponentially damped in the directions perpendicular to the interface [1]. Furthermore, SPPs could be used to transfer optical information in a two-dimensional (2D) environment. This appealing property can be used for optical addressing of different 2D optical systems and nanostructures located at a dielectric/metal interface. Actually several 2D SPP devices including passive nanostructures including mirrors or beam splitter and active elements like molecules or quantum dots are currently under development and investigation. Developments such as these raise the prospect of a new branch of photonics using SPPs, for which the term “plasmonics” emerged [2–4].

However, for experimental investigations of optical devices an important characteristic of SPP modes is that their spatial extent is governed and defined by the geometry of the nanoelements rather that by the optical wavelength [7]. This consequently opens possibilities for breaking the diffraction limit but requires instruments of observation adapted essentially to the subwavelength regime and being capable of imaging the propagation of SPPs in their 2D environment. Usually the analysis of the subwavelength regime implies necessarily near field optical (NFO) methods [5,6] able to collect the evanescent (i.e., non-radiative) components of the electromagnetic fields associated with SPPs. However, when the metal film on which the 2D optical elements are built is thin enough (i.e., with a thickness below 80–100 nm) and when the subtratum optical constant (usually glass) is higher than the one of the superstratum medium an other possibility for analyzing SPP propagation occurs. This possibility is based on the detection of coherent leaking of SPPs through the substratum. Such a far-field optical method is called leakage radiation microscopy (LRM) [8–10] and allows indeed a direct quantitative imaging and analysis of SPP propagation on thin metal films.

The aim of this article is to present a short overview of recent progress in the field of SPP imaging using LRM. In a first part of this work we will describe the theoretical principles underlying LRM. In the second part we will discuss modern leakage radiation methods and illustrate the LRM potentialities by analyzing few experiments with SPP waves interacting with 2D plasmonic devices.

2. Leakage radiation and surface plasmon polaritons

In order to describe the theoretical mechanisms explaining leakage radiation it will be sufficient for the present purpose to

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limit our analysis to the case of a metal film of complex permittivity $\epsilon_1(\omega) = \epsilon_1' + i\epsilon_1''$ ($\omega = 2\pi c/\lambda$ is the pulsation) sandwiched between two dielectric media of permittivity ϵ_0 (substrate) and $\epsilon_2 < \epsilon_0$ (superstratum). This system is theoretically simple and to a good extent experimentally accessible [1,11]. In the limiting case where the film thickness D is much bigger than the SPP penetration length in the metal (i.e., $D \gtrsim 70$ nm for gold or silver in the visible domain) one can treat the problem as two uncoupled single interfaces. We will consider as an example the interface 0/1 (the media 0 and 1 are located in the domain $z \geq 0$ and $z \leq 0$ respectively). Such an interface will be identified in the following with the plane $z =$ const. in cartesian coordinates. An elementary harmonic SPP wave is actually a TM electromagnetic mode characterized by its pulsation ω and its magnetic field $\mathbf{H} = [0, H_y, 0]$ where the y component can be written

$$\begin{aligned} H_0 &= \alpha \mathrm{e}^{\mathrm{i}k_x x}\,\mathrm{e}^{\mathrm{i}k_{z0} z}\,\mathrm{e}^{-\mathrm{i}\omega t} \quad \text{in the medium } 0 \\ H_1 &= \mathrm{e}^{\mathrm{i}k_x x}\,\mathrm{e}^{\mathrm{i}k_{z1} z}\,\mathrm{e}^{-\mathrm{i}\omega t} \quad \text{in the medium } 1, \end{aligned} \tag{1}$$

and where $k_x = k_x' + ik_x''$ is the (complex valued) wave vector of the SPP propagating in the x direction along the interface. $k_{zj} \equiv k_j = \pm\sqrt{[(\omega/c)^2\epsilon_i - k_x^2]}$ are the wave vectors in the medium $j = [0(\text{dielectric}), 1(\text{metal})]$ along the direction z normal to the interface. By applying boundary conditions to Maxwell's equations one deduces additionally $\alpha = 1$ and

$$\frac{k_1}{\epsilon_1} - \frac{k_0}{\epsilon_0} = 0, \tag{2}$$

which implies

$$k_x = \pm\left(\frac{\omega}{c}\right)\sqrt{\frac{\epsilon_0\epsilon_1}{\epsilon_0 + \epsilon_1}} \tag{3}$$

$$k_j = \pm\left(\frac{\omega}{c}\right)\sqrt{\frac{\epsilon_j^2}{\epsilon_0 + \epsilon_1}} \tag{4}$$

for a SPP wave propagating along the x direction. The choice of the sign convention connecting the z and x components of the wave vector is a priori arbitrary and must be done only on a physical ground. Indeed, due to ohmic losses in the metal we expect an exponentially decaying SPP wave propagating along the interface. This condition implies the relation $k_x' \cdot k_x'' \geq 0$ [11]. This inequality is actually always fulfilled since from Eq. (3) one deduces

$$k_x' \cdot k_x'' = \frac{1}{2}\left(\frac{\omega}{c}\right)^2 \frac{\epsilon_0^2\epsilon_1''}{(\epsilon_0 + \epsilon_1')^2 + (\epsilon_1'')^2} > 0 \tag{5}$$

which is indeed positive because $\epsilon_1'' > 0$. By writing $k_{zj} = k_j' + ik_j''$ one additionally obtains the relation

$$-k_0' \cdot k_0'' = \frac{(\omega/c)^2\epsilon_1''}{2} - k_1' \cdot k_1'' = k_x' \cdot k_x'' \geq 0. \tag{6}$$

This relation fixes the sign conventions since the wave must also decay exponentially when going away from the interface in both media. More precisely one gets

$$k_0' \cdot k_0'' \leq 0, \tag{7}$$

$$k_1' \cdot k_1'' =\geq 0. \tag{8}$$

The product $k_1' \cdot k_1''$ is positive if $\epsilon_1' \geq -|\epsilon_1|^2/(2\epsilon_0)$, a fact which is indeed true for silver and gold interfaces with air or glass in most of the visible optical domain. However small negative values of Eq. (8) occur for silver close to the interband region around $\lambda \sim 350$ nm. Additionally a higher value of ϵ_0 will also change the sign in Eq. (8). Fig. 1 shows the behavior of the SPP magnetic field close to an interface gold/air and gold/glass at the optical wavelength $\lambda = 800$ nm. At such a wavelength the conditions given by Eqs. (5)–(8) impose the solutions

$$k_x = \pm\left(\frac{\omega}{c}\right)\sqrt{\frac{\epsilon_0\epsilon_1}{\epsilon_0 + \epsilon_1}}, \quad k_j = -\left(\frac{\omega}{c}\right)\sqrt{\frac{\epsilon_j^2}{\epsilon_0 + \epsilon_1}}. \tag{9}$$

The real parts of the k_z components of the SPP wave vector are for both media oriented in the same direction corresponding to a wave propagating from the air side to the metal side (see inset in Fig. 1). Furthermore the waves are exponentially damped when going away from the interface in agreement with Eq. (7) and (8) (see Fig. 1). Most important for us is that the Poynting vector [12]$\mathbf{S} = \text{Real}[\mathbf{E} \times \mathbf{H}^*]/2$ is defined in the medium j by

$$\mathbf{S}_j = \frac{1}{2}c\,\text{Real}\left[\frac{k_x\hat{\mathbf{x}} + k_j\hat{\mathbf{z}}}{\omega\epsilon_j/c}\right]\mathrm{e}^{-2k_x''x - 2k_z''z}. \tag{10}$$

On the dielectric side the energy flow is as expected oriented in the direction of Real[$\mathbf{k}$]. However it can be shown on the metal side and for wavelengths not too close from the spectral region associated with the interband transition of gold or silver that the energy flow in the x direction is oriented oppositely to the wave vector Real[k_x] since $\text{Real}[k_x/\epsilon_1] = (k_x'\epsilon_1' + k_x''\epsilon_1'')/|\epsilon_1|^2$ is dominated by $k_x' \cdot \epsilon_1'$ and since $\epsilon_1' < 0$. However

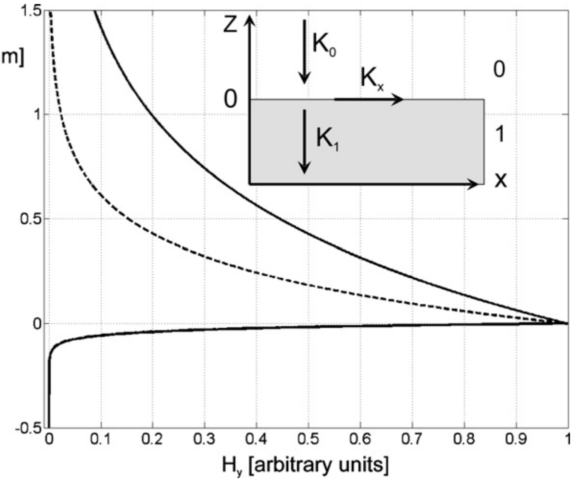

Fig. 1. Structure of the SPP magnetic field Real[H_y] across an interface air/gold (thick line) and glass/gold (dashed line). The optical wavelength considered is $\lambda = 800$ nm. The permittivity of glass is taken to be $\epsilon_{\text{glass}} = 2.25$. The inset shows the conventions for the axes x and z. The arrows indicate the direction of the real part of the wave vector normally and parallely to the interface.