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Unraveling the relationship between electromagnetic field intensity and the magnetic modulation of the wave vector of coupled surface plasmon polaritons

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Abstract. The magnetic-field-induced wave-vector modulation of surface plasmon polaritons (SPPs) is analytically derived for dielectric/metal and dielectric/metal/dielectric systems when a very thin magneto-optical metallic film is placed at different positions in the metal. In the simplest case of a single dielectric/metal interface, the SPP wave-vector modulation is found to be proportional to the intensity of the electromagnetic field at the location of the magneto-optically active layer. For the more complex dielectric/metal/dielectric systems, the SPPs existing at each dielectric/metal interface interact to give rise to modes with a symmetric- and anti-symmetric-like character. We show that in this case the relationship between wave-vector modulation and field distribution is more complex and does not follow a proportionality law for coupled eigenmodes.

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1. Introduction

Surface plasmons (SPs) are electromagnetic (EM) excitations that are spatially confined at dielectric/metal interfaces as a result of collective oscillations of the electron plasma. These SPs can be seen as propagating waves in continuous metallic layers [1], called surface plasmon polaritons (SPPs). SPPs have found a variety of applications covering the development of biosensors [2, 3], enhancement of optical transmission through nanoporous films [4], or miniaturized optical devices such as optical circuits [5] or plasmonic interferometers [6–8]. A further step in the development of optical circuitry is to gain control of the plasmon properties by means of an external agent. There have been several attempts at using agents such as temperature [9, 10], EM waves [11], electric fields [12, 13] or magnetic fields [14, 15]. In this work we will restrict ourselves to the study of the case in which this external agent is a static magnetic field, thus exploiting the magneto-optical (MO) effect. For a review on magneto-plasmonics, see [16]. In the optical range of wavelengths, the MO effect can be fairly well represented by the presence of nonzero off-diagonal elements in the dielectric tensor [17]. A good magneto-plasmonic system is a well-balanced hybrid system presenting a smart combination of plasmonic properties and MO activity. This can be achieved in two ways: one considering a ferromagnetic dielectric and a good plasmonic metal [18, 19], the other endorsing MO activity to the metal and keeping the dielectric ‘passive’. The second path, the one to be explicitly considered here, requires some material engineering. Metals with good plasmonic properties, such as gold or silver, present extremely small off-diagonal elements to have noticeable effects for typical laboratory magnetic fields (∼1 T) [20]. On the other hand, ferromagnetic metals exhibit off-diagonal elements proportional to the magnetization [17] and therefore large MO activity but they also present high absorption, making the associated SPPs difficult to employ in most practical applications. One way of overcoming these issues is to consider metal combinations such as, for instance, a Au/Co/Au trilayer [21–25]. When an external magnetic field is applied parallel to the trilayer plane and perpendicular to the SPP propagation direction, there is a nonreciprocal variation of the SPP wave vector $k$ upon magnetization reversal, i.e. $k(M) \neq k(-M)$, $M$ being the magnetization of the Co layer:

$$\frac{\Delta k}{k} \equiv \frac{k(M) - k(-M)}{k(0)}.$$  

(1)

One interesting aspect of using plasmonic resonances is that their excitation provides a high localization of the EM field at the interface between the metal and the dielectric. Thus,
many studies have found a correlation between the MO effect and the intensity of the EM field at the location or the MO material [26, 27]. The goal of this work is to study in a fundamental way the relationship between wave-vector modulation and the intensity of the SPP EM field within the MO material in trilayered magneto-plasmonic metals. From a practical point of view, a knowledge of the role played by the EM field distribution would allow the finding of routes to enhance the modulation and thereby have greater control of SPP propagation. Specifically, we analyze the SPP wave-vector modulation in flat dielectric/metal (D–M) and dielectric/metal/dielectric (D–M–D) systems when a very thin film of ferromagnetic (thus MO) metal is buried inside a noble metal, in terms of its thickness, distance to the interface and EM field distribution within it. For D–M systems we will establish that there is almost a one-to-one correlation between the wave-vector modulation and the EM field intensity in the MO layer. On the other hand, we will show that such correspondence is not so direct for interacting plasmons in D–M–D systems, where symmetric (SM)- and anti-symmetric (AM)-like eigenmodes are formed. In this case, the wave-vector modulation will be proportional not to the EM field intensity of the coupled mode in the MO layer but to the weighted difference of the EM field distribution at the MO layer of the equivalent noninteracting modes localized at each interface, from which the eigenmode originates.

This paper is organized as follows. First we will revisit the basic equations needed to perform an analytic treatment for SPPs in multilayer films including MO materials, by considering a single interface, i.e. a semi-infinite dielectric in contact with a semi-infinite, homogeneous, MO active metal (section 2). Although this is well known, it will serve to fix conventions and define some quantities. Then, in section 3 we will analyze the SPP wave-vector modulation when the semi-infinite MO metal is replaced by a noble metal with a thin film of MO material embedded in its interior. In section 4 we will extend the analysis to the case where the metallic trilayer lies between two semi-infinite dielectrics, and the total thickness is small enough to allow coupling between the two plasmon polaritons. We will consider the case of identical and different dielectrics, before concluding with a summary.

2. Single-interface dielectric/MO metal

SPP modes are well understood within a framework of classical Maxwell’s equations and arise when considering evanescent solutions (i.e. the EM field decays exponentially at both sides of the interface). The simplest geometry sustaining these modes is that of a single flat interface between a dielectric and a metal. Naming \( \varepsilon_d \) and \( \varepsilon_m \) as the permittivity of the dielectric and the metal, respectively, it follows that \( \varepsilon_d > 0 \) and \( \text{Re}(\varepsilon_m) < 0 \) is necessary to ensure that the SPP is confined to the interface, which is only possible for transverse magnetic (TM) polarization [28].

For this TM mode, continuity of the electric and magnetic in-plane components at the interface, and the dispersion relation in both media, leads to the well-known SPP dispersion relation

\[
k_{\text{spp}} = k_0 \sqrt{\frac{\varepsilon_d \varepsilon_m}{\varepsilon_d + \varepsilon_m}},
\]

where \( k \) is the in-plane component of the SPP wave vector in the absence of the MO effect and \( k_0 \) is the vacuum wave vector (\( k_0 = c/\omega \)). If the whole metal presents MO activity and a static, external magnetic field is applied, the dispersion relation of the SPP wave vector is changed. It is important to consider in which direction the magnetic field is applied to modulate the wave vector without inducing radiation losses due to TM–transverse electric polarization.
conversion, so preserving the nature of the SPP mode. The appropriate configuration is the so-called transverse MO Kerr effect (TMOKE), where the magnetic field is applied along the sample plane and perpendicular to the propagation direction. If we consider the $xy$-plane for the sample and the plasmon propagation direction along the $x$-direction, the magnetic field $B$ (enough to saturate the sample) must be applied along the $y$-direction, and the corresponding dielectric tensor for the MO active material can be written as

$$
\varepsilon_{\text{MO}}(\pm M_y) = \begin{pmatrix}
\varepsilon_{xx} & 0 & \pm M_y \varepsilon_{xz} \\
0 & \varepsilon_{xx} & 0 \\
\mp M_y \varepsilon_{xz} & 0 & \varepsilon_{xx}
\end{pmatrix},
$$

where $\varepsilon_{xz}$ is the tensor element responsible for MO activity and $M_y$ is the magnetization normalized to its saturation value ($0 \leq M_y \leq 1$).

In that case, the wave vector of an SPP propagating along the $x$-direction is given by

$$
k(\pm M_y) = k_{\text{spp}} \sqrt{\alpha \pm \beta \varepsilon_{xz}},
$$

where $\alpha$ and $\beta$ are two functions insensitive to the magnetization direction since their dependence is quadratic with the magnetic field. In the most usual cases, $|\varepsilon_{xz}| \ll |\varepsilon_{xx}|$ so the wave-vector modulation

$$
\Delta k \equiv k(\pm M_y) - k(\mp M_y) \text{ can be taken to first order in } \varepsilon_{xz},
$$

$$
\Delta k = 2k_0 M_y \varepsilon_{xz} \frac{\varepsilon_d^2}{\varepsilon_d^2 - \varepsilon_{xx}^2 \varepsilon_{xz} + \varepsilon_d} + O(\varepsilon_{xz}^2).
$$

It is important to note that the above expression is odd under mirror symmetry with respect to the interface plane. This means that, preserving the propagation direction of the SPP and the sign convention of the magnetic field, a switch of the metal/dielectric relative positions makes $\Delta k$ change its sign.

### 3. Surface plasmon polariton (SPP) wave-vector modulation in trilayered D–M systems

Let us then begin our study with a refined configuration, used in several studies previously [8, 29, 30], consisting of a very thin layer of MO material (thickness $\delta$) embedded in a semi-infinite noble metal layer, which is put in contact with a semi-infinite dielectric layer (see the scheme in figure 1). In that configuration the relevant quantities are the distance of the MO layer to the interface $d$, and the respective dielectric functions (or tensors) $\varepsilon_{\text{MO}}$, equation (3), for the MO layer, $\varepsilon_m$ for the host metal and $\varepsilon_d$ for the dielectric. To calculate the wave-vector modulation, we consider the following regions I ($z > 0$), II ($0 > z > -d$), III ($-d > z > -d - \delta$) and IV ($-d - \delta > z$), and the spatial distribution of the EM field of the SPP in each of them. The magnetic fields are described as

$$
H = H_y = A_1 e^{-i\delta z} e^{ikx},
$$

$$
H = H_y = (B_2 e^{i\alpha z} + A_2 e^{-i\alpha z}) e^{ikx},
$$

$$
H = H_y = (B_3 e^{i\beta_{\text{MO}} z} + A_3 e^{-i\beta_{\text{MO}} z}) e^{ikx},
$$

$$
H = H_y = B_4 e^{i\delta z} e^{ikx}.
$$

The associated electric fields can be obtained through Maxwell’s equations. The constants appearing on the exponentials in the $z$-direction are related through their respective dispersion relations ($\det[k \otimes k - (k \cdot k) I_{3 \times 3} + k_0 \varepsilon] = 0$) in each medium that, to first order in $\varepsilon_{xz}$, lead to
\[ k^2 = \varepsilon_d^2 + k_0^2 \varepsilon_d = \varepsilon_m^2 + k_0^2 \varepsilon_m = \varepsilon_{\text{MO}}^2 + k_0^2 \varepsilon_{\text{MO}}. \]  

In order to have plasmonic behavior, it is required that \( \text{Re}(\xi_d).\text{Re}(\xi_m) > 0 \). Next we apply continuity of the in-plane components of the fields (E, H) at each interface (\( z = 0, z = -d, z = -d - \delta \)), which leads to a 6 \times 6 system of homogeneous linear equations (\( q \equiv \varepsilon_{zz}/\varepsilon_{xx} \)),

\[
\begin{pmatrix}
1 & -1 & 0 & 0 & 0 & 0 \\
0 & 0 & 1 & -1 & 0 & 0 \\
0 & 0 & 0 & 0 & 1 & -1 \\
i\varepsilon_d \varepsilon_m & i\varepsilon_m \varepsilon_d & -i\varepsilon_m \varepsilon_d & 0 & 0 & 0 \\
e^{-\varepsilon_m d} & -e^{-\varepsilon_m d} & e^{-\varepsilon_m d} & 0 & 0 & 0 \\
i\varepsilon_m \varepsilon_d e^{-\varepsilon_m d} & i\varepsilon_m \varepsilon_d e^{-\varepsilon_m d} & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0
\end{pmatrix} \begin{pmatrix}
A_1 \\
B_2 \\
A_2 \\
B_3 \\
A_3 \\
B_4
\end{pmatrix} = \begin{pmatrix}
0 \\
0 \\
0 \\
0 \\
0 \\
0
\end{pmatrix}.
\]

A nontrivial solution requires that the determinant of the coefficient matrix must vanish. If \( \delta \) is small enough, we can consider that \( e^{\pm \varepsilon_m (d + \delta)} = e^{\pm \varepsilon_m d} (1 \pm \varepsilon_m \delta) \), \( e^{\pm \varepsilon_m (d + \delta)} = e^{\pm \varepsilon_m d} (1 \pm \varepsilon_m \delta) \). After some simple mathematical manipulation, one arrives at the following expression:

\[
k(\pm) = k_{\delta=0} \pm 2i \delta k_0^2 \frac{M_s \varepsilon_{zz}}{\varepsilon_{xx}} \frac{\varepsilon_m^2 \varepsilon_d^2}{(\varepsilon_m + \varepsilon_d)^2 (\varepsilon_m - \varepsilon_d)} e^{2 \varepsilon_m z} + O(\varepsilon_{zz}^2/\varepsilon_{xx}^2),
\]

\( k_{\delta=0} \) being given by equation (2).

Figure 1 shows (open circles) the real part of the wave-vector modulation \( \Delta k/k \) (normalized to \( \Delta k/k(d = 0) \), i.e. when the thin MO metal is located right at the interface with the dielectric semi-infinite layer) due to the presence of a 1 nm thick cobalt layer (\( \delta = 1 \)) buried at different depths in gold. The continuous line is the intensity of the EM field for a cobalt-free air/gold plasmon (also normalized to the value at the D–M interface). The dielectric constants used for calculations are \( \varepsilon_{xx} = -14.14 + i 21.49, \varepsilon_{zz} = 0.98 + i 0.15, \varepsilon_m = -15.41 + i 0.65 \). These values have been taken from experimental data of gold and cobalt at a wavelength of 700 nm.
Figure 2. Deviation of wave-vector modulation from linearity. (a) Open circles represent the relative wave-vector modulation \(\text{Re}(\Delta k/\text{Re}(k))\) for a cobalt film embedded 10 nm within the gold layer as a function of cobalt thickness. The straight, dashed line corresponds to the thin film limit. (b) Deviation from linearity as a function of depth and thickness in percentage.

From equations (5)–(8) we can see that the SPP EM field intensity in the metal has a decay factor \(\exp[2\text{Re}(\xi_m)z]\), which is exactly the same as the wave-vector modulation appearing in equation (10), which explains the agreement between the two quantities.

One important aspect is the validity of equation (10) as the thickness of the MO layer increases. Figure 2(a) shows (open circles) the exact calculation (using a transfer matrix formalism [18]) of the wave-vector modulation caused by a film, of different thicknesses, of cobalt located 10 nm from the metal/dielectric interface. The dashed line represents the ‘thin film limit modulation’, i.e. the modulation given by equation (10) considering the thin film located at the same position. A noticeable deviation of the ideal thin limit can be seen for thicknesses above 3 nm. It should be noted that this value depends on the depth of the film within the metal. Figure 2(b) shows the deviation from the thin film limit as a function of both depth and thickness. This deviation is numerically defined as \(1 - (\Delta k_{\text{Exact}}/\Delta k_{\text{Thin Film}})\times 100\), in %. As we can see, the deviation increases as a function of thickness, but it is larger when the film position is nearer the interface.

4. SPP wave-vector modulation in trilayered D–M–D systems

Let us now consider the case of a metal layer with dielectric constant \(\varepsilon_a\) limited by two dielectrics with identical dielectric constant \(\varepsilon_d\). For a very thick metal layer, the structure is able to sustain two independent SPP modes, SPP1 and SPP2, each one located at each interface (see the sketch in figure 3(a)), and which we assume propagate in the same direction along the \(x\)-axis. These modes do not interact, and thus if a thin MO layer is introduced close to the interface of SPP1 in the presence of a magnetic field, the wave vector of SPP2 will not be affected and the wave-vector modulation will be similar to that analyzed in section 2, i.e. exponential decay related to interface distance (open circles in figure 3(a)). Conversely, if the thin MO layer is introduced close to the interface of SPP2, the wave vector of SPP1 will not be affected and the wave-vector modulation will be the same in magnitude but with reversed sign, as explained at the end of section 1 (open squares in figure 3(a)).

Neglecting the effect of the MO thin film for a moment, as the thickness of the metal layer becomes smaller, the SPP1 and SPP2 modes start to interact giving rise to the well-known
Figure 3. (a) Normalized wave-vector modulation (open circles and squares) and normalized EM field intensity distribution in the metal (continuous and dashed lines) of two independent SPP modes (SPP1 and SPP2). (b) The same (open circles) for the SM SPP mode. The continuous line represents the difference of EM intensities of SPP1 and SPP2 modes. The inset shows the total EM field intensity of the SM mode. (c) As in (b) but for the AM mode. The inset shows the total EM field intensity in the metal layer.

SM and AM modes (see the sketches in figures 3(b) and (c)), which have been extensively studied [1, 28]. For each SM/AM mode, the EM field (magnetic part) intensity inside the metal layer \((z = 0\) at the SPP1 interface and \(z = -T\) at the SPP2 interface) can be expressed as

\[
H_\pm^z = H_y^\pm = (A_{\text{SPP1}}^\pm e^{z_\pm z} + A_{\text{SPP2}}^\pm e^{-z_\pm(T+z)}) e^{ik_{\pm x}} = H_{\text{SPP1}}^\pm + H_{\text{SPP2}}^\pm, \tag{11}
\]

where \(A_{\text{SPP1}}^\pm, A_{\text{SPP2}}^\pm\) are adequate amplitude constants and \(z_\pm\) satisfy the dispersion relations \(\tanh(z_\pm T) = -\frac{\xi_m^\pm}{\xi_m^\pm + \xi_d}\). The plus/minus sign refers to SM/AM modes, respectively. The decay constants \(z_\pm\) are linked with SM/AM wave-vector modulation through \(k_{\pm z}^2 = (\xi_m^\pm)^2 + k_0^2\). It is important to mention that SPP1 and SPP2 result for the actual geometry (finite size metal, not for semi-infinite conditions) and that they are different for the SM and AM modes since their characteristic wave vectors are different. Unlike the previous section, when we consider the thin MO layer inside the metal, it is not possible to find an explicit expression for
Figure 4. (a) Normalized (with respect to the SPP1 interface) wave-vector modulation (open circles) for LIM. The continuous line represents the difference of EM intensities of SPP1 and SPP2 modes whereas the inset shows the total EM field intensity of the mode. (b) As in (a) but for the HIM, normalized in this case with respect to the SPP2 interface. The inset shows again the total EM field intensity in the metal layer.

Wave-vector modulation even for small thicknesses and small MO values. One would then be left to use symmetry arguments to link aspects of the modulation with those arising from the spatial distribution of the EM field.

Taking into account the well-known profiles for SM and AS modes in standard IMIs (reproduced in the insets of figures 3(b) and (c), normalized to the intensity at the SPP1 interface), one would expect the modulation to follow them and thus be symmetric and anti-symmetric, respectively. The calculated wave-vector modulation shows quite different behavior, however. For instance, from figure 3(b) it is easy to deduce that the wave-vector modulation for the SM mode is not proportional to the EM field within the MO film: the wave-vector modulation (again, normalized to the value at the SPP1 interface) goes to zero when the MO thin layer is in the middle of the metallic layer, but the EM field in the middle presents a finite value. Interestingly, a similar profile is obtained for the AM mode. Notice that here we are dealing with normalized values with respect to the SPP1 interface; the actual values of the modulation and of the EM field are different for the SM and AM cases.

The exhibited profiles lead us to think that the modulation can be regarded as a combination of ‘incoherent’ SPPs propagating in the same direction, but in opposite interfaces (thus, with opposite sign). This argument leads us to propose that the wave-vector modulation should have the form

\[ \left( \frac{\Delta k}{k} \right)_+ \sim A_{SPP1}^+ e^{-i k_{d1} d} - A_{SPP2}^+ e^{-i k_{d2} (T-d)}, \]

\[ \left( \frac{\Delta k}{k} \right)_- \sim A_{SPP1}^- e^{-i k_{d1} d} - A_{SPP2}^- e^{-i k_{d2} (T-d)}, \]

i.e. proportional to the difference of the weighted EM field distribution of SPP1 and SPP2 evaluated at the MO layer location. The relative minus sign highlights the opposite behavior
of each SPP modulation with respect to the field direction, as mentioned at the end of section 2. To verify our assumption, we calculated (see figure 3) the normalized values of the wave-vector modulation (open circles) and of the result of equation (12) (continuous line) for a 1 nm cobalt thin film at different positions within a 100 nm gold layer surrounded by air at both interfaces, for both the symmetric (figure 3(b)) and anti-symmetric (figure 3(c)) SPP modes. The schemes at the left of the figures show the behavior of SM and AM modes. As can be observed, the results show very good agreement, confirming the ‘incoherence’ hypothesis.

This idea can be further verified by exploring a geometrically asymmetric situation when different dielectrics are located at each side of the metallic layer, e.g. \( \varepsilon_d \) for \( z > 0 \), \( \varepsilon_D \) for \( z < -T \) (see the sketches in figures 4(a) and (b)). In this situation there are no longer SM and AM modes, but lower index (LIM) and higher index (HIM) modes, respectively. Figure 4 shows the same magnitudes as in figure 3 for the LIM and HIM modes, respectively, for \( \varepsilon_d = 1 \) and \( \varepsilon_D = 1.01 \). The schemes again capture the behavior of the LIM and HIM modes. As can be observed, the zero crossing is not right at the center of the metal layer, but slightly displaced (60 and 40 nm for the LIM and HIM modes, respectively), reflecting the geometrical asymmetry.

5. Conclusions

Summarizing, we have presented a semi-analytic study of the relationship between wave-vector modulation and EM field distribution in SPPs for dielectric/metal and dielectric/metal/dielectric systems when a thin MO film is placed at different positions inside a noble metal thicker layer. For dielectric/metal systems the wave-vector modulation due to the MO active thin layer is proportional to the amount of EM field inside the MO layer, this proportionality being linear for thin enough films. For dielectric/metal/dielectric systems, where the equivalent SPP modes existing at each side of the metallic layer give rise to the well-known SM/AM (or LIM/HIM) modes, we find that the wave-vector modulation depends on the weighted difference of the EM field distribution of the equivalent noninteracting, or incoherent, SPP1 and SPP2 modes at the MO layer.

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