Properties of surface plasmon polaritons on lossy materials: lifetimes, periods and excitation conditions

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Abstract
The possibility to excite surface plasmon polaritons (SPPs) at the interface between two media depends on the optical properties of both media and geometrical aspects. Specific conditions allowing the coupling of light with a plasmon-active interface must be satisfied. Plasmonic effects are well described in noble metals where the imaginary part of the dielectric permittivity is often neglected (‘perfect medium approximation (PMA)’). However, some systems exist for which such approximation cannot be applied, hence requiring a refinement of the common SPP theory. In this context, several properties of SPPs such as excitation conditions, period of the electromagnetic field modulation and SPP lifetime then may strongly deviate from that of the PMA. In this paper, calculations taking into account the imaginary part of the dielectric permittivities are presented. The model identifies analytical terms which should not be neglected in the mathematical description of SPPs on lossy materials. These calculations are applied to numerous material combinations resulting in a prediction of the corresponding SPP features. A list of plasmon-active interfaces is provided along with a quantification of the above mentioned SPP properties in the regime where the PMA is not applicable.

Keywords: plasmon lifetime, surface plasmon polaritons, lossy material

(Some figures may appear in colour only in the online journal)

1. Introduction

Surface plasmon polaritons (SPPs) are collective oscillations of electrons occurring at the interface of materials. More than hundred years after their discovery [1], SPPs have promoted new applications in many fields such as microelectronics [2], photovoltaics [3], near-field sensing [4–6], laser technology [7, 8], photonics [9, 10], meta-materials design [11], high order harmonics generation [12], or charged particles acceleration [13, 14].

Most of these applications are based on expensive noble metals such as gold, silver or platinum, as these materials greatly support the plasmonic phenomena, exhibit very small (plasmonic) losses and the experimental results match well with the associated theory [1, 4, 7, 15, 16]. Although there were numerous studies addressing SPPs in lossy materials [17–29], some specific aspects remain to be investigated.

In this paper, a mathematical condition for SPP excitation at flat interfaces is provided. This approach includes the widely accepted theory but reveals a wider (material dependent) domain of SPP excitation than predicted by the existing literature. The importance of the terms originating from losses is underlined and complemented by formula of the SPP near-field period and lifetime.

2. Excitation conditions for SPPs in lossy materials

At a planar interface between two different materials, the electric field components ($E_{x,y,z}$) and magnetic field...
were taken positive here, accounting for the complex wavenumber of the SPPs propagating along the surface normal axis are indicated as which surface plasmon polaritons are considered. The wavevector of medium (Equation 1) of medium $i$ by $\varepsilon_i = n_i^2$, and the complex-valued wave-numbers $k_i \in \mathbb{C}$ (associated with electromagnetic field modes in the medium $i$). At the interface between two media (1 and 2), the conservation of light momentum results in the condition

$$k_{1,2}^2 = \beta^2 - k_0^2 \varepsilon_{1,2},$$

(1)

where $\beta \in \mathbb{C}$ is the SPP wavenumber along the interface, $k_0 = \frac{\omega}{c}$ is the wavenumber of the incident light ($\omega \in \mathbb{R}$: light angular frequency, $c$: light velocity in vacuum).

In TM geometry, the continuity conditions for the electromagnetic fields results in the relation [1, 16, 30]

$$\frac{k_1}{\varepsilon_1} + \frac{k_2}{\varepsilon_2} = 0.$$  

(2)

Equation (2) represents the dispersion relation of SPPs at the interface between two semi-infinite media. The signs of $\Re(k_1)$ and $\Re(k_2)$ were taken positive here, accounting for the exponential decay of the electromagnetic field amplitude in the direction perpendicular to (away from) the interface. The combination of equations (1) and (2) provides the solutions of the SPP wavenumber $\beta$

$$\beta = \pm \frac{\omega}{c} \sqrt{\frac{\varepsilon_1 \varepsilon_2}{\varepsilon_1 + \varepsilon_2}}.$$  

(3)

In order to address the mathematical description of damped surface electromagnetic waves, in the following, we selected the positive branch of $\beta$.

It must noted that in a generalized view, the characterization of lossy waves can be treated by calculating an observable response function $F(\omega, \beta)$ which allows to construct a dispersion relation by locating its complex zeros/ poles [21, 23].

As already noted by Ritchie et al [23], when damping is relevant, the dispersion relation $\omega(\beta)$ for $\beta \in \mathbb{R}$ may have complex solutions ($\omega \in \mathbb{C}$). Conversely, if $\omega$ is real-valued, $\beta$ may be complex-valued. Although straightforward in a theoretical framework, there is some ambiguity about the significance of complex values of $\omega$ or $\beta$ in the interpretation of experiments [23].

In experiments it may be difficult to observe temporal or spatial decay of a resonance due to its rapidity or smallness. The properties of such excitations are usually extracted from the transfer of energy and momentum to the system, involving both real $\omega$ and real $k$. As an example, dispersion relations have been determined by attenuated total reflection in Otto configuration [4]. In this approach, a beam is totally reflected at the basis plane of an optical prism. Excitation of SPP in a neighborhood metal surface may be realized via coupling through a gap of a dielectric medium (air). SPP manifest as drops in the totally reflected signal, when momentum matching between light and SPP occurs. Experimentally, as underlined by Kovener et al [21], this can be realized either for a variation of frequency $\omega$ at a fixed angle of incidence $\theta$, or via a variation of $\theta$ at a fixed $\omega$. The first procedure produces dispersion curves with a specific ‘bend back’ feature, while the second procedure results in curves without that feature [21].

To calculate the excitation conditions of SPPs at the interface between two arbitrary (absorbing) media, a mathematical analysis is presented in the following.

2.1. Sign analysis on the dispersion relation

SPPs can be excited only if the dispersion relation (equation (2)) is fulfilled. In order to extract the SPP excitation conditions from the dispersion relation, a sign analysis can be performed on the real and imaginary parts of equation (2), which can be mathematically developed to:

$$\begin{cases}
\Re \left( \frac{k_1}{\varepsilon_1} + \frac{k_2}{\varepsilon_2} \right) = 0, \\
\Im \left( \frac{k_1}{\varepsilon_1} + \frac{k_2}{\varepsilon_2} \right) = 0.
\end{cases}$$

(4)

By assuming $\Re(k_1) > 0$ and $\Re(k_2) > 0$,

this equation can be used to deduce a constraint on the sign of real part of the dielectric permittivities $\varepsilon_{1,2}$, resulting in

$$\Re \left( \frac{\varepsilon_1}{\varepsilon_2} \right) < 0.$$  

(5)

Equation (5) defines the first necessary condition for excitation of SPPs, which is equivalent to

$$\Re(\varepsilon_1) \times \Re(\varepsilon_2) + \Im(\varepsilon_1) \times \Im(\varepsilon_2) < 0.$$  

(6)

important for lossy materials

The physical meaning of equation (6), named Condition 1 for SPP excitation, is the following: in presence of a perfect dielectric medium (e.g. for $\Im(\varepsilon_1) = 0$ or $\Im(\varepsilon_2) = 0$), equation (6) implies that SPPs can only be excited at a dielectric–metal interface. This is usually fulfilled when $\Im(\varepsilon_{1,2}) \ll |\Re(\varepsilon_{1,2})|$. Then equation (6) reduces to the..
widely accepted expression [1, 30, 31]
\[
\Re(\varepsilon_1) \times \Re(\varepsilon_2) < 0. 
\] (7)

However, in presence of two absorbing media (having then \(3m(\varepsilon_1) > 0\) and \(3m(\varepsilon_2) > 0\), the physical meaning is less intuitive. The condition given by equation (6) is more complex due to non-vanishing contributions of the imaginary parts of the dielectric permittivities. Consequences of this additional term, important for lossy materials, will be discussed in section 2.3.

2.2. Perfect medium approximation (PMA) and beyond

Assuming purely real-valued dielectric permittivities, equation (3) is typically used to derive another condition for SPP excitation. This approach is called PMA and will be outlined in the following. For simplicity, we adopt the following notations: \(\varepsilon'_i = \Re(\varepsilon_i)\) and \(\varepsilon''_i = \Im(\varepsilon_i)\). If \(\varepsilon'_1 = 0\) and \(\varepsilon''_2 = 0\), then \(\beta\) is real-valued (\(\Re(\beta)\)), and equation (3) becomes
\[
\begin{align*}
\Re\left(\frac{\varepsilon_1\varepsilon_2}{\varepsilon_1 + \varepsilon_2}\right) & > 0, \\
3m\left(\frac{\varepsilon_1\varepsilon_2}{\varepsilon_1 + \varepsilon_2}\right) & = 0.
\end{align*}
\] (8)

The latter can be rewritten as
\[
\begin{align*}
\left(\varepsilon'_1\varepsilon'_2 - \varepsilon''_1\varepsilon''_2\right) + \left(\varepsilon'_1\varepsilon''_2 + \varepsilon'_2\varepsilon''_1\right) & > 0, \\
\left(\varepsilon'_1 + \varepsilon'_2\right)^2 + \left(\varepsilon''_1 + \varepsilon''_2\right)^2 & > 0, \\
\left(\varepsilon'_1 + \varepsilon'_2\right)^2 + \left(\varepsilon''_1 + \varepsilon''_2\right)^2 & = 0,
\end{align*}
\]
which is strictly equivalent to
\[
\frac{\varepsilon'_1\varepsilon'_2}{\varepsilon'_1 + \varepsilon'_2} > 0.
\] (9)

Condition (equation (9)) implies
\[
|\varepsilon'_2| > \varepsilon'_1
\] (10)
and is widely used in literature [16, 31]. In particular, when medium 1 is air (\(\varepsilon_1 = 1\)), the joint application of equations (7) and (10) leads to the well admitted condition
\[
\Re(\varepsilon_2) < -1.
\] (11)

However, beyond the PMA, it must be noted that equation (3) can be treated using fully complex permittivity values since \(\beta\) is defined in \(C\) for any value of the dielectric permittivities \(\varepsilon_i\). As a consequence, in presence of one (or more) ‘lossy’ materials, e.g., when \(3m(\varepsilon_1) > 0\) or \(3m(\varepsilon_2) > 0\), there is no other restriction for SPP excitation than the Condition 1 given by equation (6). In other words, performing an ad hoc restriction of the dielectric permittivity to its real part(s) may lead to an oversimplified SPP excitation condition.

2.3. Exploration of plasmon-active material combinations

In this section, the plasmon activity of a wide set of material combinations is explored by comparing predictions of the PMA (real-valued equations (7) and (9)) with the more general case (complex-valued equation (6)). For that, if not stated differently, data of the optical constants were taken from [32]. Values are listed in the appendix.

In a first step, a selection of different metals exposed to air are analyzed. The results for the SPP-activity for 12 different noble and transition metals are provided in table 1. Here we restrict the study to two wavelengths frequently used in laser processing: 800 nm and 400 nm. However, these calculations can be generalized to other material combinations and wavelengths. The SPP-activity of the metal–air interfaces is indicated by a tick (√), whereas the interfaces which do not support SPPs are marked by a cross (×). At both wavelengths the generalized model (equation (6)) predicts similar results as the PMA. Interestingly, and opposed to the PMA, niobium (Nb) is predicted to support SPPs when interfaced with air, upon 400 nm irradiation.

In a second step, the same metals in contact with a semi-infinite dielectric medium (SiO2) were addressed. Table 2 lists the corresponding results on SPP-activity. Again, with a few exceptions, agreement is found between the PMA and the generalized approach. The case of Nb is similar as in the analysis for air (compare tables 1 and 2). Interestingly, at 400 nm wavelength, the PMA predicts no SPP-activity for the SiO2/Au interface, while the more general treatment does. Also for SiO2-covered Cr at 800 nm wavelength, the predictions of both theories do not match.

In a third step, three most relevant metals (Ag, Au, Ti) in contact with a dielectric (air, Al2O3, SiO2, TiO2, ZnO) or semiconducting (GaAs, GaP, Ge, InP, Si, SiC) medium were analyzed regarding their SPP-activity at 800 and 400 nm wavelengths. Table 3 compiles the corresponding information and indicates that for Au and Ag at 800 nm, both models provide the same predictions on SPP-activity. However, for Ti at 800 nm and for all three metals at 400 nm, differences between predictions of both models can be observed. For Ti, the differences between those models arise for the significant contribution of the imaginary part of the dielectric permittivity (see equation (6), \(\varepsilon'_2(\text{Ti}, \lambda = 800 \text{ nm}) = -2.85 + i19.11, \varepsilon'_2(\text{Ti}, \lambda = 400 \text{ nm}) = -2.22 + i6.66\)). For Au and Ag at 400 nm wavelength, the different model predictions arise from a similar origin (see table A1).

3. Period of SPPs in lossy materials

3.1. Modeling the plasmon period

The spatial period \(\Lambda\) of the electromagnetic field can be calculated from the complex-valued SPP wavenumber \(\beta\) (using equation (3)) via [17, 33]
\[
\Lambda = \frac{2\pi}{\Re(\beta)}
\] (12)
with
\[
\Re(\beta) = \frac{\omega}{2c} \sqrt{2[F_1^2 + F_2^2]^{1/2} + 2F_1},
\]
Table 1. Analysis of SPP-activity of several metal ($\varepsilon_2$) surfaces irradiated in air ($\varepsilon_1 = 1$) at $\lambda = 800$ nm and 400 nm wavelengths. Meaning of the symbols: $\checkmark$: SPP are excited, $\times$: SPP are not excited at the interface. Bold font indicates interfaces where the prediction deviates from accepted theories.

| Interface | $\lambda = 800$ nm | $\lambda = 400$ nm |
|-----------|-----------------|-----------------|
| $3m(\varepsilon_2) = 0$ | $3m(\varepsilon_2) = 0$ | $3m(\varepsilon_2) = 0$ |
| Air/Ag    | $\checkmark$    | $\checkmark$    | $\checkmark$    |
| Air/Al    | $\checkmark$    | $\checkmark$    | $\checkmark$    |
| Air/Au    | $\checkmark$    | $\checkmark$    | $\checkmark$    |
| Air/Cr    | $\checkmark$    | $\checkmark$    | $\checkmark$    |
| Air/Cu    | $\checkmark$    | $\checkmark$    | $\checkmark$    |
| Air/Fe    | $\checkmark$    | $\checkmark$    | $\checkmark$    |
| Air/Nb    | $\checkmark$    | $\checkmark$    | $\times$        |
| Air/Ni    | $\checkmark$    | $\checkmark$    | $\checkmark$    |
| Air/Pt    | $\checkmark$    | $\checkmark$    | $\checkmark$    |
| Air/Ti    | $\checkmark$    | $\checkmark$    | $\checkmark$    |
| Air/V     | $\times$        | $\times$        | $\checkmark$    |
| Air/W     | $\times$        | $\times$        | $\times$        |

Table 2. Analysis of SPP-activity of several metal surfaces ($\varepsilon_2$) irradiated at $\lambda = 800$ nm and 400 nm wavelengths through SiO$_2$ as covering medium [$\\varepsilon_2(\\text{SiO}_2, \lambda = 800 \text{ nm}) = 2.11$, $\\varepsilon_2(\\text{SiO}_2, \lambda = 400 \text{ nm}) = 2.16$]. Meaning of the symbols: $\checkmark$: SPP are excited, $\times$: SPP are not excited at the interface. Bold font indicates interfaces where the prediction deviates from accepted theories.

| Interface | $\lambda = 800$ nm | $\lambda = 400$ nm |
|-----------|-----------------|-----------------|
| $3m(\varepsilon_2) = 0$ | $3m(\varepsilon_2) = 0$ | $3m(\varepsilon_2) = 0$ |
| SiO$_2$/Ag | $\checkmark$    | $\checkmark$    | $\checkmark$    |
| SiO$_2$/Al | $\checkmark$    | $\checkmark$    | $\checkmark$    |
| SiO$_2$/Au | $\checkmark$    | $\checkmark$    | $\times$        |
| SiO$_2$/Cr | $\checkmark$    | $\checkmark$    | $\checkmark$    |
| SiO$_2$/Cu | $\checkmark$    | $\checkmark$    | $\checkmark$    |
| SiO$_2$/Fe | $\checkmark$    | $\checkmark$    | $\checkmark$    |
| SiO$_2$/Nb | $\times$        | $\times$        | $\checkmark$    |
| SiO$_2$/Ni | $\checkmark$    | $\checkmark$    | $\checkmark$    |
| SiO$_2$/Pt | $\checkmark$    | $\checkmark$    | $\checkmark$    |
| SiO$_2$/Ti | $\checkmark$    | $\checkmark$    | $\checkmark$    |
| SiO$_2$/V  | $\times$        | $\times$        | $\checkmark$    |
| SiO$_2$/W  | $\times$        | $\times$        | $\times$        |

where

$F_1 = (\varepsilon_1'\varepsilon_2' - \varepsilon_1''\varepsilon_2'')(\varepsilon_1' + \varepsilon_2')F_0 + (\varepsilon_1''\varepsilon_2' + \varepsilon_1'\varepsilon_2'')(\varepsilon_1'' + \varepsilon_2'')F_0,$

$F_2 = (\varepsilon_1''\varepsilon_2' + \varepsilon_1'\varepsilon_2'')(\varepsilon_1' + \varepsilon_2')F_0 - (\varepsilon_1''\varepsilon_2' - \varepsilon_1'\varepsilon_2'')(\varepsilon_1'' + \varepsilon_2'')F_0,$

and $F_0 = [(\varepsilon_1' + \varepsilon_2')^2 + (\varepsilon_1'' + \varepsilon_2'')^2]^{-1}$. The terms underlined with a curly brackets can be neglected if the PMA is used, i.e., if $\varepsilon_1'' = 0$ and $\varepsilon_2'' = 0$. From equation (12), it is obvious that the imaginary parts of both media can significantly affect the SPP periods.

For the PMA ($\varepsilon_1'' = 0$ and $\varepsilon_2'' = 0$), equation (12) reduces to the expression [31]

$$
\lambda_{\text{PMA}} = \frac{2\pi}{\Re(\beta)} = \frac{\lambda}{\sqrt{\varepsilon_1 + \varepsilon_2}}.
$$

However, it should be underlined that equation (13) should not be applied to lossy materials.

### 3.2. Exploration of plasmon-active material combinations

In this section, different SPP-active combinations of materials are systematically explored in terms of their SPP period using equation (12).

Figure 2 shows for metals being SPP-active in air the spatial period $\Lambda$ as a function of their real parts of $\varepsilon_2$. At 800 nm wavelength, the SPP-periods calculated by equation (12) (discrete data points) match within 5% to the irradiation wavelength $\lambda$—as indicated by a horizontal red dashed line. The solution based on the PMA (equation (13)) with $\varepsilon_1 = 1$ is shown as red solid line. By comparison, it is obvious that the PMA (equation (13)) should not be applied to lossy materials. At 400 nm wavelength, the SPP-periods more strongly depend on the material. A reduction of $\Lambda$ by up to 25%, when compared to the wavelength $\lambda$, is observed for Ag, Li, and Na —see the blue horizontal dashed line, with a similar mismatch to the PMA curve (blue solid line).
Table 3. Analysis of SPP-activity of three metal surfaces (Ag, Au, Ti) irradiated through different dielectric or semiconducting media at \( \lambda = 800 \) and 400 nm wavelengths. Meaning of the symbols: \( \checkmark \): SPP are excited, \( \times \): SPP are not excited at the interface. Bold font indicates interfaces where the prediction deviates from accepted theories.

| Interface     | \( \lambda = 800 \) nm |          | \( \lambda = 400 \) nm |          |
|---------------|-------------------------|----------|-------------------------|----------|
|               | \( 3m(e_2) = 0 \)      |          | \( 3m(e_2) = 0 \)      |          |
| Air/Ag        | \checkmark              | \checkmark| \checkmark              | \checkmark|
| \( \text{Al}_2\text{O}_3/\text{Ag} \) | \checkmark              |          | \checkmark              |          |
| \( \text{GaAs} /\text{Ag} \) | \checkmark              |          | \checkmark              |          |
| \( \text{GaP}/\text{Ag} \) | \checkmark              |          | \checkmark              |          |
| \( \text{Ge}/\text{Ag} \) | \checkmark              |          | \checkmark              |          |
| \( \text{InP}/\text{Ag} \) | \checkmark              |          | \checkmark              |          |
| \( \text{Si}/\text{Ag} \) | \checkmark              |          | \checkmark              |          |
| \( \text{SiC}/\text{Ag} \) | \checkmark              |          | \checkmark              |          |
| \( \text{SiO}_2/\text{Ag} \) | \checkmark              |          | \checkmark              |          |
| \( \text{TiO}_2/\text{Ag} \) | \checkmark              |          | \checkmark              |          |
| \( \text{ZnO}/\text{Ag} \) | \checkmark              |          | \checkmark              |          |
| Air/Au        | \checkmark              |          | \checkmark              |          |
| \( \text{Al}_2\text{O}_3/\text{Au} \) | \checkmark              |          | \checkmark              |          |
| \( \text{GaAs}/\text{Au} \) | \checkmark              |          | \checkmark              |          |
| \( \text{GaP}/\text{Au} \) | \checkmark              |          | \checkmark              |          |
| \( \text{Ge}/\text{Au} \) | \checkmark              |          | \checkmark              |          |
| \( \text{InP}/\text{Au} \) | \checkmark              |          | \checkmark              |          |
| \( \text{Si}/\text{Au} \) | \checkmark              |          | \checkmark              |          |
| \( \text{SiC}/\text{Au} \) | \checkmark              |          | \checkmark              |          |
| \( \text{SiO}_2/\text{Au} \) | \checkmark              |          | \checkmark              |          |
| \( \text{TiO}_2/\text{Au} \) | \checkmark              |          | \checkmark              |          |
| \( \text{ZnO}/\text{Au} \) | \checkmark              |          | \checkmark              |          |
| Air/Ti        | \checkmark              |          | \checkmark              |          |
| \( \text{Al}_2\text{O}_3/\text{Ti} \) | \checkmark              |          | \checkmark              |          |
| \( \text{GaAs}/\text{Ti} \) | \( \times \)            |          | \( \times \)            |          |
| \( \text{GaP}/\text{Ti} \) | \( \times \)            |          | \( \times \)            | \checkmark|
| \( \text{Ge}/\text{Ti} \) | \( \times \)            |          | \( \times \)            |          |
| \( \text{InP}/\text{Ti} \) | \( \times \)            |          | \( \times \)            |          |
| \( \text{Si}/\text{Ti} \) | \( \times \)            |          | \( \times \)            |          |
| \( \text{SiC}/\text{Ti} \) | \( \times \)            |          | \( \times \)            |          |
| \( \text{SiO}_2/\text{Ti} \) | \( \times \)            |          | \( \times \)            |          |
| \( \text{TiO}_2/\text{Ti} \) | \( \times \)            |          | \( \times \)            | \checkmark|
| \( \text{ZnO}/\text{Ti} \) | \checkmark              |          | \checkmark              |          |

Figure 3 shows the results of analogous analyses for SPP-active metals (\( e_2 \)) in contact with \( \text{SiO}_2 \) (\( \varepsilon \)). At 800 nm wavelength, the SPP-periods match within 8% to the wavelength in \( \text{SiO}_2 \) (i.e., above the metal surface, \( \lambda/\Re\sqrt{\varepsilon} \)) [34] —as indicated by a horizontal red dashed line. At 400 nm wavelength, for most metals, the deviations from the latter expression also stay below 10% (compared to the horizontal blue dashed line). In contrast, for Ag, Li and Na, larger differences up to 65% are evident. For both wavelengths, the data points and the periods calculated using the PMA (equation (13), solid lines) match only for specific materials.

In order to understand the role of the overlayer, the SPP-period is investigated at the interface between Au (\( e_2 \)) and various semi-transparent materials (\( \varepsilon \)). The results are provided in figure 4. They indicate that, again, the SPP-periods strongly depends on \( \varepsilon \). Specifically, a reasonably good agreement is found between the SPP-period \( \Lambda \) and the wavelength in the covering medium \( \lambda/\Re\sqrt{\varepsilon} \). This can be seen by the red solid (\( \lambda = 800 \) nm) and blue dashed (\( \lambda = 400 \) nm) lines, which quantitatively agree with the data points for small values of \( \Re(\varepsilon) \). The remarkable deviations at larger values of \( \Re(\varepsilon) \) arise from the PMA.

Figure 5 shows analogous results for the case of a Ti substrate (\( e_2 \)) covered by the same set of materials (\( \varepsilon \)). Again, the lines represent the wavelengths in the covering medium \( \lambda/\Re\sqrt{\varepsilon} \), which are in reasonable agreement with the data points.

It should be underlined that the conclusions drawn in this work strongly depend on the quality of the optical data used. All parameters affecting the dielectric permittivities may influence the predictions, i.e., the temperature [35], high-intensity illumination [36–38], strong external fields, etc. Moreover, we have assumed that the SPP-active interface is surrounded by two semi-infinite half-spaces. This assumption may break down when additional interfaces fall within the vertical extent of SPP field. Then another model has to be
used which considers the coherent coupling of SPP at several interfaces [20, 39].

4. Lifetime of SPPs in lossy materials

So far, we have described the SPPs within the frame of steady-state conditions. Once the driving external radiation field is turned off, the SPPs may exist for a certain duration called lifetime $\tau_{SPP}$ in the following. Upon internal damping, via electron collisions, re-radiation of light to the far field [40] via scattering on defects, etc., SPP typically vanish on a sub-picosecond timescale. However, the exact lifetime strongly depends on the material and irradiation parameters.

In this section, two different models of SPP lifetime are introduced and applied to the materials Ag and Ti in a wide range of irradiation wavelengths (from UV to IR). Two different models from literature will be compared [16, 41].

The SPP propagation length $L_{SPP}$ (1/e-decay of the intensity) is defined by [41]

$$L_{SPP} := [2 \times 3m(\beta)]^{-1}. \quad (14)$$

Using the group velocity ($v_g$) and the lifetime ($\tau_{SPP}$) of SPPs, their propagation length can be approximated by:

$$L_{SPP} \sim v_g \times \tau_{SPP}. \quad (15)$$
The group velocity is given by the definition
\[ v_g = \frac{\text{d} \omega}{\text{d}[\text{Re}(\beta)]} = c \frac{\text{Re}(\tau_{\text{SP}}) - \lambda \text{Im}(\omega)}{\text{Im}(\omega)} \]
with \( \text{Re}(\tau_{\text{SP}}) \).

Equations (14)–(16) result in an SPP lifetime of
\[
\tau_{\text{SP}} \approx \frac{1}{2 \times \text{Im}(\omega_{\text{SPP}}) \times v_g}.
\]

Another expression of the SPP lifetime was provided by Raether [16] to be:
\[
\tau_{\text{SP}} = \frac{1}{2 \times \text{Im}(\omega_{\text{SPP}})},
\]
where
\[
\text{Im}(\omega_{\text{SPP}}) = \frac{c}{2} \frac{\text{Re}(\beta) \text{Im}(\varepsilon_2)}{\text{Re}(\varepsilon_2)^2} + \frac{\text{Re}(\varepsilon_1) \text{Re}(\varepsilon_2)}{\text{Re}(\varepsilon_1) + \text{Re}(\varepsilon_2)}.
\]

In the following two subsections, we apply and compare the two equations (17) and (18) to the well-known case of air–Ag interface (example 1, a material properly described by the PMA) and the less studied air–Ti interface (example 2, representing a lossy material).

**Example 1 (lossless material): air–Ag interface**

Figure 6 presents the SPP lifetime \( \tau_{\text{SP}} \) as a function of the irradiation wavelength \( \lambda \) (350 nm–2 \( \mu \)m). The two curves shown in figure 6 are based on equations (17) and (18) along with the optical data from [43]. Both curves exhibit an excellent agreement featuring lifetimes up to 6 ps. At 400 nm wavelength, \( \tau_{\text{SP}} \) accounts to 14 fs (equation (18)) and 27 fs (equation (17)), while at 800 nm wavelength, both equations provide a very similar value of 1 ps. At 400 nm wavelength, \( \tau_{\text{SP}} \) agrees reasonably well with the experiments of Kubo et al [44] who observed the SPP decay within 50 fs for the air–Ag interface. It should be underlined that the SPP lifetime strongly depends on the irradiation wavelength.

**Example 2 (lossy material): air–Ti interface**

Figure 7 displays the SPP lifetime \( \tau_{\text{SP}} \) at the air–Ti interface, as a function of the irradiation wavelength \( \lambda \). Again, the two curves shown in figure 7 are calculated from equations (17) and (18) using the optical data from [43]. In contrast to Example 1 (Ag), the two curves are remarkably different for the air–Ti interface. The model based on equation (18) provides values which are smaller by more than one order of magnitude when compared to the ones provided by equation (17). Specifically, at 400 nm wavelength, \( \tau_{\text{SP}} \) accounts to 0.3 fs (equation (18)) versus 3 fs (equation (17)). At 800 nm wavelength, it accounts to 0.5 fs (equation (18)) versus 11 fs (equation (17)). The values calculated by equation (18) appear unreasonably small here, as they are smaller than the expected Drude relaxation time of the electrons [45]. It is important to emphasize that the two models result in very different predictions for Ti.

### 5. Conclusion

In this work, the properties of SPPs at the interface between two lossy materials have been studied. Our mathematical analysis allowed to identify terms which should not be neglected in the mathematical description of SPPs on lossy materials. This rigorous approach was applied to numerous material combinations (dielectric/metal, semiconductor/metal), providing a generalized criterion for SPP excitation along with the quantification of the SPP periods. For Ag and Ti surfaces interfaced with air, the lifetimes of the SPP were reported in a wide spectral range between 350 nm and 2 \( \mu \)m.
Table A1. Summary of the dielectric permittivities used in this work.

| Material | Wavelength | Reference |
|----------|-------------|-----------|
|          | 800 nm      | 400 nm    |
| Air      | 1.00        | 1.00      | —         |
| Ag       | −27.95      | 3.52      | 3.77      | 0.67 | [43] |
| Al2O3    | 3.10        | 3.19      | 3.19      | —    | [32] |
| Al       | −63.55      | 47.31     | 23.39     | 4.77 | [32] |
| Au       | −26.15      | 1.85      | −1.08     | 6.49 | [32] |
| Be       | 0.17        | 23.22     | −1.42     | 18.11| [32] |
| Bi       | −6.61       | 21.89     | −2.32     | 6.76 | [46] |
| a-C      | 4.80        | 3.37      | 3.53      | 3.77 | [32] |
| Co       | −17.65      | 25.20     | −7.74     | 7.63 | [32] |
| Cr       | −1.42       | 36.29     | −10.65    | 10.76| [32] |
| Cu       | −25.27      | 2.52      | −3.49     | 5.22 | [32] |
| Diamond  | 5.77        | 0.00      | 6.08      | 0.00 | [32] |
| Ethanol  | 1.84        | 0.00      | —         | —    | [47] |
| Fe       | −6.38       | 20.19     | −3.05     | 8.27 | [32] |
| GaAs     | 13.55       | 0.63      | 14.53     | 18.76| [32] |
| GaP      | 10.13       | 0.00      | 17.53     | 2.31 | [32] |
| Ge       | 21.96       | 3.01      | 12.24     | 18.34| [32] |
| Graphite | 5.95        | 11.58     | 5.20      | 6.76 | [32] |
| H2O      | 1.76        | 0.00      | 1.82      | 0.00 | [32] |
| InP      | 11.93       | 1.46      | 16.49     | 15.30| [32] |
| Li       | −14.25      | 2.16      | −2.58     | 0.96 | [32] |
| Mo       | 2.08        | 24.52     | −1.19     | 19.91| [32] |
| Mg       | −58.35      | 11.33     | −12.41    | 1.25 | [46] |
| Na       | −11.84      | 0.35      | −2.30     | 0.21 | [32] |
| Nb       | −6.74       | 14.49     | −0.61     | 12.90| [32] |
| Ni       | −13.04      | 21.73     | −2.98     | 7.60 | [32] |
| Pt       | −16.47      | 28.11     | −5.11     | 9.77 | [32] |
| a-Si     | 15.17       | 0.85      | 13.39     | 18.84| [32] |
| Si       | 13.63       | 0.048     | 30.85     | 4.30 | [32] |
| SiC      | 6.78        | 0.00      | 7.64      | 0.00 | [32] |
| SiO2     | 2.11        | 0.00      | 2.16      | 0.00 | [32] |
| Ta       | −11.17      | 7.90      | 2.49      | 12.60| [32] |
| Ti       | −6.21       | 25.2      | −4.36     | 12.60| [43] |
| TiO2     | 7.78        | 0.00      | 11.55     | 0.00 | [32] |
| V        | 1.88        | 21.76     | −4.92     | 17.24| [32] |
| W        | 5.22        | 19.44     | 5.68      | 16.34| [32] |
| ZnO      | 3.80        | 0.00      | 5.11      | 0.07 | [48] |
| Zr       | −10.56      | 16.38     | −2.81     | 7.18 | [49] |

wavelength pointing out again that plasmonic loss mechanisms should be carefully taken into consideration.

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Appendix. Optical constants of materials

In this work, several sources of optical data were used. For Al2O3, Al, Au, Be, a-C, Co, Cr, Cu, diamond, ethanol, Fe, GaAs, GaP, Ge, graphite, H2O, InP, Li, Mo, Na, Nb, Ni, Pt, c-Si, a-Si, SiC, SiO2, Ta, TiO2, V, and W, the optical data were mostly taken from [32]. For Mg, the work of Hagemann et al was used [46], for ZnO, the work of Postava et al [32]. The detailed optical data used in this work are summarized in table A1.

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