Propagating surface plasmons 5

5.1 Surface and particle electron oscillation modes

Introductory examples

To introduce the concept of plasma oscillations, we consider a thin metallic film. The metal is described using a simple model : we assume that there are n free and independent electrons per unit volume. The crystal lattice is modelled by a uniform positively charged background. This is the so-called *jellium model*. The purpose of this section is to illustrate the essence of a plasmon as an oscillatory collective mode of the electrons. We work entirely within the framework of classical mechanics.

Let us assume that a static homogeneous electric field $\mathbf{E}_{\text{ext}} = E_{\text{ext}}\hat{x}$ is applied normally to the film along the x-axis, with $E_{\text{ext}} < 0$. A force $-e\mathbf{E}_{\text{ext}}$ is exerted on the electrons, which are displaced by a distance $x > 0$. A negative surface charge $-nex$ appears on the right interface and a positive surface charge on the left interface. These surface charges generate a static electric field that cancels the external field inside the metal.

At time $t = 0$, the external electrostatic field is switched off. The system is now out of equilibrium and free to evolve. The electrons are accelerated by the electric field generated by the surface charges. When they return to their equilibrium position, they have acquired momentum and continue moving, generating an electric field of opposite sign. This process repeats periodically, leading to collective oscillations of the electrons.

The motion of a single electron along the x-axis is governed by Newton's law

$$m\frac{d^2x}{dt^2} = -eE_x, \tag{5.1}$$

where magnetic forces and dissipative mechanisms have been neglected.

Using Gauss's theorem, the electric field generated by a sheet carrying a surface charge density nex is $nex/(2\varepsilon_0)\,\mathbf{u}$, where $\mathbf{u}$ is the outward unit normal. Accounting for the two interfaces of the film, the resulting electric field inside the metal is

$$E_x = \frac{nex}{\varepsilon_0}. \tag{5.2}$$

Equation (5.1) then becomes

$$\frac{d^2x}{dt^2} + \frac{ne^2}{m\varepsilon_0}x = 0. \tag{5.3}$$

This equation allows us to introduce the plasma frequency ω_p, defined by

$$\omega_p^2 = \frac{ne^2}{m\varepsilon_0}. \tag{5.4}$$

The plasma frequency corresponds to the natural frequency of collective oscillations of the electron gas in the bulk of the metal. The oscillation is produced by the electric field generated by all electrons, which explains why it is referred to as a *collective* oscillation. This simple argument captures the essence of a plasmon.

As discussed in the chapter on near-field physics, a resonator corresponds to a periodic exchange of energy between two forms of energy : kinetic and potential energy for a mass attached to a spring, electric and magnetic energy for an LC circuit or an optical microcavity. A plasmonic system corresponds to a periodic exchange between the electric energy stored in the surface charges (the system behaves as a capacitor) and the kinetic energy of the electrons. Unlike photonic cavities, where inertia is provided by magnetic energy, here it originates from the kinetic energy of the electrons. This is the key mechanism enabling electromagnetic energy confinement in subwavelength volumes beyond the diffraction limit.

For noble metals, the electronic density places the plasma frequency in the near ultraviolet or visible spectral range, while highly doped semiconductors can sustain plasmonic oscillations in the mid-infrared. Plasmonic resonances therefore enable the fabrication of nanoscale resonators at optical frequencies.

Surface plasmon of a thin film

If we consider the electric field generated by a single interface, the field amplitude is reduced by a factor of two. The equation of motion becomes

$$\frac{d^2x}{dt^2} + \frac{ne^2}{2m\varepsilon_0}x = 0, \tag{5.5}$$

leading to a resonance frequency

$$\omega = \frac{\omega_p}{\sqrt{2}}. \tag{5.6}$$

Plasmon resonance of a nanosphere

We now consider a metallic nanosphere much smaller than the wavelength, so that retardation effects can be neglected. The electrostatic field generated by a uniform polarization **P** inside a sphere is given by

$$\mathbf{E} = -\frac{\mathbf{P}}{3\varepsilon_0}. \tag{5.7}$$

Using Newton's equation,

$$m\frac{d^2x}{dt^2} = -eE_x, \tag{5.8}$$

and noting that the polarization results from electron displacement,

$$P_x = -nex, \tag{5.9}$$

we obtain

$$\frac{d^2x}{dt^2} + \frac{ne^2}{3m\varepsilon_0}x = 0. \tag{5.10}$$

The resonance frequency of the plasmon in a nanosphere is therefore

$$\omega = \frac{\omega_p}{\sqrt{3}}. \tag{5.11}$$

This result illustrates that the plasmon resonance frequency depends strongly on the geometry, similarly to mechanical resonators such as tuning forks or organ pipes.

5.2 Surface electromagnetic wave

We now consider modes that propagate along an interface and are confined in the vicinity of that interface. More precisely, we seek solutions that decay exponentially away from the interface. In this sense, a single interface acts as a waveguide.

At this stage, no specific assumption is made regarding the nature of the materials. Hence, the dispersion relation derived below applies to any material (e.g. metals, dielectrics) and any frequency range (e.g. radio waves, infrared, visible). When surface waves propagate at metal–dielectric interfaces at infrared or optical frequencies, they are referred to as surface plasmon polaritons (SPPs). Other surface waves exist, such as surface phonon polaritons in the infrared or THz range, and surface radio waves at metal–dielectric interfaces.

The only assumptions made in the following are that the media are local, isotropic, and non-magnetic. They are therefore characterized by complex, frequency-dependent dielectric constants $\varepsilon_r(\omega)$. The upper medium ($z > 0$) is labelled medium 1, and the lower medium ($z < 0$) is labelled medium 2. The wavevector is denoted by $\mathbf{k} = (k_x, k_y, \gamma)$, and its modulus by k.

Dispersion relation for the non-magnetic case

We look for source-free solutions of Maxwell's equations. In each medium $i = 1,2$, the electric field satisfies the Helmholtz equation

$$\nabla^2\mathbf{E}_i + \varepsilon_i\frac{\omega^2}{c^2}\mathbf{E}_i = 0. \tag{5.12}$$

We consider a p-polarized (transverse magnetic, TM) solution and seek fields of the form (in Chapter 6, k_x was denoted by β) :

$$\begin{aligned} z > 0: \quad E_{x1} &= E_0\, e^{ik_x x + i\gamma_1 z}, \\ z < 0: \quad E_{x2} &= E_0\, e^{ik_x x - i\gamma_2 z}, \end{aligned} \tag{5.13}$$

which ensures continuity of the tangential field at the interface. The decay constants are defined as

$$\gamma_1 = \left(\varepsilon_1 \frac{\omega^2}{c^2} - k_x^2\right)^{1/2}, \qquad \mathrm{Im}(\gamma_1) > 0, \tag{5.14}$$

$$\gamma_2 = \left(\varepsilon_2 \frac{\omega^2}{c^2} - k_x^2\right)^{1/2}, \qquad \mathrm{Im}(\gamma_2) > 0, \tag{5.15}$$

so that the fields decay exponentially away from the interface.

We seek transverse waves, meaning that $\nabla \cdot \mathbf{E} = 0$ in each medium. In Fourier space, this condition becomes

$$\mathbf{k} \cdot \mathbf{E} = 0, \tag{5.16}$$

with $\mathbf{k} = (k_x,0,\gamma)$. Since $\mathbf{k}$ is complex, this relation should not be interpreted as a geometric orthogonality condition. Transverse ($\nabla \cdot \mathbf{E} = 0$) should therefore not be confused with perpendicular.

The z-components of the electric field are then

$$\begin{aligned} z > 0: \quad E_{z1} &= -\frac{k_x}{\gamma_1} E_0\, e^{ik_x x + i\gamma_1 z}, \\ z < 0: \quad E_{z2} &= \frac{k_x}{\gamma_2} E_0\, e^{ik_x x - i\gamma_2 z}. \end{aligned} \tag{5.17}$$

Imposing the continuity of the normal component of $\varepsilon \mathbf{E}$ at the interface yields

$$\varepsilon_1 \gamma_2 = -\varepsilon_2 \gamma_1. \tag{5.18}$$

Equation (5.18) is the dispersion relation of the surface mode. Squaring both sides (which introduces an unphysical second branch that must be discarded later), one obtains an explicit relation between frequency and wavevector. The solution of interest for TM polarization is

$$k_{\mathrm{sp}}^2(\omega) = \frac{\omega^2}{c^2} \frac{\varepsilon_1 \varepsilon_2}{\varepsilon_1 + \varepsilon_2}. \tag{5.19}$$

Equation (5.19) has two branches corresponding to $\varepsilon_1 \gamma_2 = \pm \varepsilon_2 \gamma_1$. Only the solution with the minus sign corresponds to a physical surface mode.

At this stage, it is not yet obvious that Eq. (5.19) indeed describes a surface-confined wave. Let us assume that medium 1 is a lossless dielectric with real, positive ε_1, and that medium 2 is lossless with $\mathrm{Im}(\varepsilon_2) = 0$. To ensure exponential decay in medium 1, we require

$$k_{\mathrm{sp}} > \sqrt{\varepsilon_1}\,\frac{\omega}{c}. \tag{5.20}$$

From Eq. (5.19), this condition implies

$$\frac{\varepsilon_2}{\varepsilon_1 + \varepsilon_2} > 1, \tag{5.21}$$

which can only be satisfied if

$$\varepsilon_1 + \varepsilon_2 < 0. \tag{5.22}$$

As a consequence, γ_2 is purely imaginary, ensuring exponential decay in medium 2 as well. We have thus identified a surface electromagnetic wave for TM polarization.

A similar surface mode can exist for transverse-electric (TE) polarization in magnetic media with permeabilities μ_i. The dispersion relation then reads

$$\mu_1\gamma_2 = -\mu_2\gamma_1. \tag{5.23}$$

Link with resonances of the reflection factor

An alternative way to derive the surface-mode dispersion relation is to examine the poles of the Fresnel reflection coefficients. Since the reflected field can be written as

$$E_r^{s,p} = r_{s,p} E_{\mathrm{inc}}^{s,p}, \tag{5.24}$$

the Fresnel reflection coefficient $r_{s,p}$ can be interpreted as a linear response function to the incident field, viewed as an external excitation. As for any linear system, a resonance corresponds to a pole of this response function.

Thus, setting the denominator of $r_{s,p}$ equal to zero yields the dispersion relation of the surface mode. For TM polarization in non-magnetic media, the denominator of the Fresnel coefficient is proportional to $\varepsilon_1\gamma_2 + \varepsilon_2\gamma_1$, recovering Eq. (5.18).

For magnetic media, the Fresnel reflection coefficients can be written as

$$r_s = \frac{\mu_2\gamma_1 - \mu_1\gamma_2}{\mu_2\gamma_1 + \mu_1\gamma_2}, \qquad r_p = \frac{\mu_1\varepsilon_2\gamma_1 - \mu_2\varepsilon_1\gamma_2}{\mu_1\varepsilon_2\gamma_1 + \mu_2\varepsilon_1\gamma_2}. \tag{5.25}$$

The corresponding dispersion relations are therefore

$$s\text{-polarization}: \quad \mu_2\gamma_1 + \mu_1\gamma_2 = 0, \qquad p\text{-polarization}: \quad \mu_1\varepsilon_2\gamma_1 + \mu_2\varepsilon_1\gamma_2 = 0. \tag{5.26}$$

This approach is particularly useful for more complex geometries such as thin-film stacks, where it naturally accounts for guided modes, interface modes, and coupling between them.

A technical remark is in order. The Fresnel coefficients are often expressed in terms of the incident angle θ_i rather than the parallel wavevector component k_x. For a propagating incident wave in a lossless dielectric with refractive index n_1, one may use $k_x = n_1(\omega/c)\sin\theta_i$. However, for surface waves, we seek real values of k_x larger than $n_1\omega/c$, so that the angle-based notation becomes ill-suited.

5.3 Surface plasmon polaritons

In this section, we consider the specific case of surface modes propagating at the interface between a metal and a dielectric in the optical regime. As mentioned in the introduction, these surface waves are called surface plasmon polaritons (SPPs). For brevity, they are often simply referred to as *surface plasmons*. The metals most commonly supporting surface plasmons are noble metals such as gold and silver.

However, it is important to stress that a wide class of materials can sustain surface plasmons, including doped semiconductors, transparent conducting oxides, and other plasmonic materials.

Dispersion relation of an SPP for a Drude model

We discuss the dispersion relation of a surface plasmon at the interface between a dielectric medium with a real dielectric constant ε_1 and a metal described by a Drude model. As already mentioned, this model provides only a crude description of noble metals near the plasma frequency but allows analytical insight.

The Drude dielectric function is written as

$$\varepsilon_2(\omega) = 1 - \frac{\omega_p^2}{\omega^2 + i\nu\omega}, \tag{5.27}$$

where ω_p is the plasma frequency and ν is the phenomenological damping rate.

Inserting this expression into the SPP dispersion relation, Eq. (13.15), yields

$$k_{\mathrm{sp}} = \frac{n_1\omega}{c}\left[\frac{\omega^2 - \omega_p^2 + i\omega\nu}{(1+\varepsilon_1)(\omega^2 + i\omega\nu) - \omega_p^2}\right]^{1/2}. \tag{5.28}$$

Because the permittivity is complex owing to absorption, it is impossible to satisfy the dispersion relation with both a real frequency and a real wavevector. This issue is regularly ecountered in nanophotonics Ther correct approach (linked to the correct definition of what a mode is) is to fix the wavevector t to be real and find solutions with a complex-valued frequency.

Quasinormal surface-plasmon modes (complex-valued frequency). When imposing a real value of k_{sp}, the dispersion relation yields a complex-valued frequency $\tilde{\omega}$. Plotting $\mathrm{Re}(\tilde{\omega})$ as a function of k_{sp}, one observes a horizontal asymptote.

Inspection of the dispersion relation shows that k_{sp} diverges when

$$\varepsilon_1 + \varepsilon_2(\omega) = 0. \tag{5.29}$$

For a lossy metal, this equation has a solution only for a complex frequency $\tilde{\omega}_{\mathrm{sp}}$. In the limit of large k_{sp}, the solution of Eq. (5.28) is

$$\tilde{\omega}_{\mathrm{sp}} = \sqrt{\frac{\omega_p^2}{\varepsilon_1 + 1}}\sqrt{1 - \frac{(\varepsilon_1 + 1)\nu^2}{4\omega_p^2}} - i\frac{\nu}{2}. \tag{5.30}$$

The associated amplitude decay time is $2/\nu$. The horizontal asymptote has two important consequences. First, it indicates that the density of quasinormal modes exhibits a peak near this frequency. Second, it implies the existence of surface modes with very large wavevectors and extremely confined fields. These modes are delocalized along the interface (varying as $\exp(ik_{\mathrm{sp}}x)$) but exhibit very low group velocity. Their decay rate is governed solely by the damping parameter ν of the metal.

Typical length scales for a propagating surface plasmon

We now consider a surface plasmon driven by a monochromatic source at real frequency ω. In the presence of losses, the SPP wavevector k_{sp} is complex. Its imaginary part accounts for the attenuation of the mode along the propagation direction.

A characteristic propagation length along the interface is defined as

$$\delta_x = \frac{1}{\mathrm{Im}(k_{\mathrm{sp}})}. \tag{5.31}$$

Two additional characteristic lengths describe the confinement of the surface plasmon perpendicular to the interface. They are defined as

$$\delta_{z_i} = \frac{1}{\mathrm{Im}(\gamma_i)}, \qquad i = 1,2, \tag{5.32}$$

where γ_i are given by Eqs. (13.11) and (13.12). Inserting the dispersion relation Eq. (13.15) yields

$$\frac{1}{\delta_{z_i}} = \mathrm{Im}(\gamma_i) = \frac{\omega}{c}\mathrm{Im}\left[\left(\frac{\varepsilon_i^2}{\varepsilon_1 + \varepsilon_2}\right)^{1/2}\right]. \tag{5.33}$$

In the visible range, the propagation length along the interface is on the order of a few micrometres, whereas in the far-infrared it can reach several tens of centimetres.

Regarding confinement, the decay length δ_{z_2} inside the metal depends only weakly on wavelength. It is mainly determined by the skin depth and is typically on the order of 10–20 nm for gold. By contrast, the decay length δ_{z_1} in the dielectric varies dramatically with frequency. For a vacuum–gold interface, it ranges from about 165 nm at $\lambda = 633$ nm to nearly 700 μm at $\lambda = 36$ μm.

These trends can be understood using the lossless Drude model $\varepsilon_2 \simeq -\omega_p^2/\omega^2$ for $\omega \ll \omega_p$. In this limit, Eq. (5.33) yields

$$\delta_{z_2} \simeq \frac{c}{\omega_p}, \tag{5.34}$$

which is non-dispersive and of the order of a few tens of nanometers. In contrast, the decay length in the dielectric can be approximated as

$$\delta_{z_1} \simeq \lambda_1 \frac{\omega_p}{\omega}, \tag{5.35}$$

where $\lambda_1 = c/(\sqrt{\varepsilon_1}\omega)$ is the wavelength in medium 1. This explains the dramatic increase of δ_{z_1} in the infrared.

The long propagation length in the far-infrared originates from the weak confinement of the mode : most of the electromagnetic energy resides in the dielectric rather than in the lossy metal. As a result, Joule losses are strongly reduced. This idea can be generalized : plasmonic losses are minimized when the modal field is predominantly located in a low-loss dielectric rather than in the metal. This principle underlies the design of hybrid metallo-dielectric structures.

Finally, we note that confinement is directly linked to the magnitude of the in-plane wavevector. Since $k_x^2 > \varepsilon_1\omega^2/c^2$, very large values of k_x lead to

$$\delta_{z_i} \simeq \frac{1}{k_x}, \tag{5.36}$$

so that modes with large wavevectors are strongly confined near the interface.

How to excite a surface plasmon

Propagating waves and surface plasmons are on different sides of the light line. At a given frequency, a propagating wave has a wavevector smaller than ω/c, whereas a surface plasmon always has a wavevector larger than ω/c. Hence, the phase cannot be the same along a plane interface and the two fields cannot satisfy boundary conditions. As a consequence, an incident plane wave on a plane interface cannot excite a surface plasmon.

The question of the excitation of a surface mode can follow different schemes. All of them need to fulfil the same condition : to generate a real parallel wavevector that matches the parallel surface-plasmon wavevector.

Point-like source. It is possible to use a point-like source. According to Weyl's expansion, the spherical wave generated by a point source contains evanescent waves with a continuum spectrum of wavevectors. Of course, these components are evanescent waves localized close to the source. Hence, the source has to be close to the surface. More generally, any subwavelength source located in close proximity to the surface can excite surface modes. It can be an atom, a tiny particle scattering an incident beam, the aperture of an elongated optical fibre, as used in near-field microscopy, a scattering tip, as used in an atomic force microscope or scanning tunnelling microscope, a tiny scratch in the surface, etc.

Kretschmann configuration. One can use a metal film with a thickness smaller than the skin depth separating two dielectric media with different dielectric constants n_1 and $n_2 > n_1$. Here, the key idea is to take advantage of a large refractive index to increase the modulus of the wavevector from $n_1\omega/c$ to $n_2\omega/c$. By illuminating from the side of the high-refractive-index medium with a plane wave with wavevector

$$k_x = n_2 \frac{\omega}{c} \sin\theta_i,$$

it is possible to excite the surface mode on the other side by taking advantage of the fact that the incidence angle can be chosen so that $k_x > n_1\omega/c$. This technique is called the *Kretschmann configuration*. Strictly speaking, with an incident plane wave of infinite spatial extent, it is the quasinormal plasmon mode that is excited; however, experimentally, beams have finite sizes and guided surface plasmons are launched.

Otto configuration. A similar but less practical technique consists in using the evanescent part of the field totally internally reflected in a prism. This is known as the *Otto configuration*. It requires accurate control of the spacing between the prism and the metal surface.

Grating excitation. A grating with a period d can be used, so that the nth order of the grating has a wavevector

$$k_{x,n} = n_1 \frac{\omega}{c} \sin\theta_i + n\frac{2\pi}{d}.$$

If the phase-matching condition

$$k_{\mathrm{sp}} = n_1 \frac{\omega}{c} \sin\theta_i + n\frac{2\pi}{d}$$

is approximately satisfied, an incident plane wave may effectively excite a surface mode. The efficiency of the coupling depends on the parameters of the surface profile of the grating. It can be optimized to allow a total absorption, as was first reported using a gold grating operating in the visible at a frequency where a flat surface is a good mirror – see T"he total absorption of light by a diffraction grating" by M.C. Hutley, D. Maystre. This is a very counterintuitive result, since the modulation depth of the sinusoidal grating is very low, so that the surface appears almost flat. This is an example of critical coupling.

5.4 Surface plasmon contributions to the LDOS

The concept of LDOS plays a key role when studying the lifetime of an emitter or the electromagnetic energy at equilibrium in the near field. Close to metallic surfaces, the LDOS is increased by orders of magnitude due to the electronic contributions.

It is known that slow-velocity systems can be used to increase the DOS. As the dispersion relation becomes flat close to the band edge, the number of states (represented by dots) with a frequency in the interval $\Delta\omega$ increases. This behaviour is known as a Van Hove singularity. The number of states provided by a surface plasmon at resonance appears infinite, as the dispersion relation becomes flat and unbounded.

The spatial dependence of the LDOS is also crucial. Modes with large parallel wavevector k decay as $\exp(-kz)$ for $k \gg k_0$. A quantitative analysis based on the imaginary part of the Green tensor yields the asymptotic expression of the normalized LDOS at a distance $z \ll \lambda$ from the interface :

$$\frac{\rho(z,\omega)}{\rho_v(\omega)} = \frac{\mathrm{Im}[\varepsilon]}{|\varepsilon+1|^2}\frac{1}{4(k_0 z)^3}. \tag{5.37}$$

The resonant surface-plasmon contribution is clearly associated with the factor $1/|\varepsilon + 1|^2$, which is responsible for a strong enhancement of the LDOS, possibly by orders of magnitude.