Introduction to Surface Plasmon Theory

Jean-Jacques Greffet

Institut d’Optique Graduate School
A  A few examples of surface plasmons

B  Surface waves
   Definition, Polarization properties, Dispersion relation, History of surface waves, Lateral wave.

C  Plasmons

D  Surface plasmon polariton (SPP)

F  Key properties of SPP

G  SPP in lossy metals

J  Fourier optics for surface plasmons
What is a surface plasmon polariton?

\[ E_0 \exp[\imath kx - \imath \gamma z - \imath \omega t] \]
Image of a SPP

Dawson PRL 94
Excitation using a sub-$\lambda$ source
Experimental proof of the existence of SPP

EELS (Electron Energy Loss Spectroscopy) of reflected electrons.

Powell, Phys.Rev. 1959
Observation of the LDOS using EELS

What is a Surface Wave (1)?

Derivation of the dispersion relation

0. Surface wave

1. Solution of a homogeneous problem

2. Pole of a reflection factor
Dispersion relation

\[ \varepsilon_2 k_{z1} + \varepsilon_1 k_{z2} = 0 \]

Reflection factor

\[ r_F = \frac{\varepsilon_2 k_{z1} - \varepsilon_1 k_{z2}}{\varepsilon_2 k_{z1} + \varepsilon_1 k_{z2}} \]

\[ K^2 = \frac{\omega^2}{c^2} \frac{\varepsilon}{\varepsilon + 1} \]

is a solution of \((\varepsilon_2 k_{z1})^2 = (\varepsilon_1 k_{z2})^2\)

Two cases: Brewster propagating wave and surface wave
What is a Surface Wave (2)?

Structure of the wave

\[ E_x \exp[ikx - i\gamma z - i\omega t] \]
1. Case of a good conductor

\[ \varepsilon_r = \frac{i \sigma}{\omega \varepsilon_0} \]

\[ k_{II} = \frac{\omega}{c} \left( 1 + \frac{i \omega \varepsilon_0}{2 \sigma} \right) \]

\[ k_z = \frac{\omega i - 1}{c \sqrt{2}} \sqrt{\frac{\omega \varepsilon_0}{\sigma}} \]
What is a Surface Wave (4)?

- Historical account of the surface wave concept
- Long radio wave propagation: the hypothesis of Zenneck
- Dipole emission above an interface: the pole contribution and the Sommerfeld surface wave.
- Norton approximate formula
- Banos contribution
- Lateral wave
What is a lateral wave?

In the **far field**, the field decay as

\[
\frac{\exp(ik_1 h + ik_2 r - ik_1 z)}{r^2}
\]

**velocity** : \(c/n_2\)

**Source**  
**Conducting medium**  
**z**  
**r**
A. Banos, Dipole radiation in the presence of a conducting half space
Pergamon Press, NY, 1966


A. Boardman Electromagnetic surface modes J. Wiley, NY 1982

R. King, Lateral electromagnetic waves, Springer Verlag, NY, 1992
Question: when a surface wave is a surface plasmon?
First example: a thin film vibrational collective mode of oscillation of electrons

\[ \omega_p^2 = \frac{n e^2}{m \varepsilon_0} \]
What is a (bulk) plasmon polariton?

Acoustic wave in an electron gas:

\[ \text{photon} + \text{phonon} = \text{polariton} \]
(Bulk) Plasmon dispersion relation

**Hydrodynamic model**

\[
\text{div } E = \frac{\rho}{\varepsilon_0}
\]

\[
\frac{\partial \rho}{\partial t} + v \cdot \nabla \rho = -\nabla P - \rho E
\]

\[
P = -\frac{\rho}{e} k_B T
\]

\[
\omega^2 = \omega_p^2 + v^2 k^2 \approx \omega_p^2
\]

**Electrodynamic point of view**

\[
\text{div } D = \text{div } \varepsilon_0 \varepsilon_r(\omega)\ E = 0
\]

\[
\varepsilon_r(\omega)\ k \cdot E(k,\omega) = 0
\]

\[
\varepsilon_r(\omega) = 1 - \frac{\omega_p^2}{\omega^2} = 0 \quad \Rightarrow \quad \omega = \omega_p
\]

An electron gas has a mechanical vibration eigenmode that generates a longitudinal EM mode. Key idea: plasmon is a material resonance.
What is a Surface Wave (2)?

Structure of the wave

\[ E_x \exp[ikx - i\gamma z - i\omega t] \]

Elliptic polarization with a (geometrically) longitudinal component. (but transverse wave)
Optical properties of a metal

Drude model

\[ \varepsilon_r(\omega) = 1 - \frac{\omega_p^2}{\omega^2 + i\omega\gamma} \]

Metal or dielectric?

- \(\omega > \omega_p\) dielectric
- \(\omega < \omega_p\) metal

Plasmon or surface wave?

- \(\omega > \gamma\) plasmon
- \(\omega < \gamma\) surface wave

\[ \varepsilon_r(\omega) = 1 - \frac{\omega_p^2}{\omega^2} \quad \text{oscillation} \]

\[ \varepsilon_r(\omega) \approx i \frac{\omega_p^2}{i\omega\gamma} \approx i \frac{\omega_p^2}{\omega\gamma} = i \frac{\sigma}{\omega\varepsilon_0} \quad \text{Overdamped oscillation} \]
Surface plasmon polariton?

\[ k = \frac{\omega}{c} \sqrt{\frac{\varepsilon}{\varepsilon + 1}} = \frac{\omega}{c} \sqrt{\frac{\omega^2 - \omega_p^2}{2\omega^2 - \omega_p^2}} \]

Remark: no surface plasmon in metals at THz frequencies
Non local correction

How good is a macroscopic analysis of the problem? What are the relevant length scales?

Definition of a non-local model
Origin of the non-locality
- Thomas Fermi screening length
- Landau Damping
Phonon polariton
Phonon polariton
Specific properties of SPP

1. Large density of states
2. Fast relaxation/broad spectrum
3. Confined fields
1. Large local density of states
Local Density of States

Energy point of view

\[ U = \frac{\omega^2}{\pi^2 c^3} \frac{1}{\exp\left(\frac{\hbar \omega}{kT}\right) - 1} \]

Lifetime point of view

\[ A_{21} = B_{21} \frac{\hbar \omega^3}{\pi^2 c^3} = \left[B_{21} \hbar \omega\right] \frac{\omega^2}{\pi^2 c^3} \]

Larger LDOS means: i) shorter lifetime, ii) larger energy at thermodynamic equilibrium
Lifetime is not intrinsic but depends on the environment

Drexhage (1970)
Chance, Prock, Silbey (1978)
Asymptotic form of the EM- LDOS

Near-field form

\[ N(\omega) = \frac{1}{16\pi^2 \omega z^3} \frac{\text{Im}[\varepsilon(\omega)]}{|1 + \varepsilon(\omega)|^2} \]

- Resonance for \( \varepsilon(\omega) \rightarrow -1 \)
- Lorentzian shape
- The near-field effect exists without SPP !!

Signature of the SPP ?

*PRL 85, 1548 (2000)*

*PRB 68, 245405 (2003)*
Where are the new modes coming from?

The EM field inherit the density of states of matter: SPP are polaritons!
Where are the *new* modes coming from?

Estimate of the number of EM states with frequency below $\omega$:

$$\frac{N}{V} = \int_{0}^{\omega} g(\omega')d\omega' = \frac{\omega^3}{3\pi^2 c^3} \quad N \approx \frac{V}{\lambda^3}$$

Estimate of the number of electrons/phonons:

$$N \approx \frac{V}{a^3}$$

The EM field inherits the large DOS of matter.
Application: nanoantenna

Greffet, Science 308 p (2005) p 1561

Kühn et al. PRL 97, 017402 (2006)

Farahani et al., PRL 95, 017402 (2005)
Anger et al., PRL 96, 113002 (2006)
Energy density close to the surface

Energy density close to the surface

\[ z = 100 \, \mu m \]

\[ z = 1 \, \mu m \]

\[ z = 100 \, nm \]

Shchegrov PRL, 85 p 1548 (2000)
Observation of the thermal near field

Application: nanoscale heat transfer

E. Rousseau, Nature Photonics, (2009), DOI 10.1038/Nphoton.2009.144
Observation of the SPP LDOS

\[ F = \int F(\mathbf{k}, \omega) \, d^3k \, d\omega \]

Remark

LDOS and projected LDOS
SPP key properties 2

Fast relaxation/Broad spectrum
Losses in noble metals (1)

Intraband loss Mechanism

Different mechanisms at high frequency and low frequency

DC-GHZ : 2 bodies interaction
optics : 3 bodies interaction
# Losses in noble metals (2)

<table>
<thead>
<tr>
<th>Collisions</th>
<th>Relaxation time</th>
</tr>
</thead>
<tbody>
<tr>
<td>Electron-phonon DC</td>
<td>$\alpha$ Te</td>
</tr>
<tr>
<td>Electron-phonon at optical frequency</td>
<td>17 fs weak dependence on Te</td>
</tr>
<tr>
<td>Electron-electron</td>
<td>170 fs</td>
</tr>
</tbody>
</table>

Applications:

- Broad spectrum antenna
- Fast hot spot
- Absorber
- Local Heater
Field confinement
Field confinement by particles, tips

Electrostatic or SPP confinement?

Examples: bow-tie, antennas, lightning rod, particles
Field confinement by particles, tips

Electrostatic or SPP confinement?

Examples: bow-tie, antennas, lightning rod, particles

Look for a resonance close to $\omega_p$. 
SPP focusing

Nano Lett., 2009, 9 (1), 235-238• DOI: 10.1021/nl802830y
Fourier optics of surface plasmons

Surface plasmon

Solution for a non-lossy medium

\[ E(z) \exp[i(K_xx + K_y y - \omega t)] \]

Dispersion relation

\[ K^2 = \frac{\omega^2}{c^2} \frac{\varepsilon}{\varepsilon + 1} \]

If \( \varepsilon \) is complex, there is no solution with real \( K \) and \( \omega \).
Surface plasmon dispersion relation

Real $\omega$ and complex $K$

Real $K$ and complex $\omega$. 
Maximum confinement of the field?

\[ \frac{1}{K_{\text{max}}} \]

No limit!
Local Density of states?

- Finite value of the LDOS
- Divergence of the LDOS
First analysis of the backbending

Analysing ATR experiments

Data taken at fixed angle while varying K

Data taken at fixed K while varying the frequency.
The field is a superposition of plane waves:

\[
\Psi(x, y, z) = \iiint \Psi(\alpha, \beta, 0) e^{i(\alpha x + \beta y + \gamma z)} \frac{d\alpha}{2\pi} \frac{d\beta}{2\pi}
\]

\[\alpha^2 + \beta^2 + \gamma^2 = \frac{\omega^2}{c^2}\]

Propagation and diffraction can be described as linear operations on the spatial spectrum.

Propagation is a low-pass filter: resolution limit.

Equivalent (Huygens-Fresnel) form:

\[
\Psi(x, y, z) = -\frac{1}{2\pi} \iiint \Psi(x', y', 0) \frac{\partial}{\partial z} \left[ \frac{\exp(ikr)}{r} \right] dx' dy'
\]
Surface plasmon Fourier optics

Brongersma, Nanolett, 2009

Hillenbrand, APL08
Key issues

Huygens-Fresnel propagator for surface plasmon ?

Linear superposition of modes with complex $K$ ?
Linear superposition of modes with complex $\omega$ ?

Implication for the maximum confinement.
Implication for the LDOS.

Link with Fourier optics.
We start from the general representation of the field generated by an arbitrary source distribution. The field is given explicitly by the Green tensor.

\[ \mathbf{E}(\mathbf{r}, t) = -\mu_0 \int dt' \int d^3 \mathbf{r}' \, \mathcal{G}(\mathbf{r}, \mathbf{r}', t - t') \frac{\partial j(\mathbf{r}', t')}{\partial t'} , \]

The Green tensor has a Fourier representation:

\[ \mathcal{G}(\mathbf{r}, \mathbf{r}', t - t') = \int \frac{d^2 K}{4\pi^2} \int \frac{d\omega}{2\pi} \mathcal{G}(\mathbf{K}, z, z', \omega) e^{i[\mathbf{K}(\mathbf{r}-\mathbf{r}') - \omega(t-t')]} , \]

It includes Fresnel reflection factor and therefore poles representing surface plasmons.
General representation of the field

Following Sommerfeld, we define the surface wave as the pole contribution to the field

\[ \vec{G} = \vec{G}_{reg} + \vec{G}_{sp}, \]

\[ \vec{E}_{sp}(\vec{r}, t) = -\mu_0 \int dt' \int d^3\vec{r}' \, \vec{G}_{sp}(\vec{r}, \vec{r}', t - t') \frac{\partial j(\vec{r}', t')}{\partial t'}. \]
General representation of the field

Evaluating the pole contribution:

We can choose to integrate either over $\omega$ or over $K_x$

\[
\vec{g}_{sp}(K, z, z', \omega) = \frac{f_{\omega \text{sp}}(K, z, z')}{\omega - \omega_{\text{sp}}} + \frac{f_{-\omega \text{sp}}(K, z, z')}{\omega + \omega_{\text{sp}}},
\]

or

\[
\vec{g}_{sp}(K, z, z', \omega) = \frac{f_{K_x \text{sp}}(K_y, z, z', \omega)}{K_x - K_{x, \text{sp}}} + \frac{f_{-K_x \text{sp}}(K_y, z, z', \omega)}{K_x + K_{x, \text{sp}}},
\]
We obtain two different representations of the SP field:

**Complex $\mathbb{K}$**

$$E = \int \frac{d\omega}{2\pi} \int \frac{dK_y}{2\pi} \left( \mathbb{K} - \frac{K_{sp}}{\gamma_m} n_m \right) E_\omega(K_y, \omega)$$

$$e^{i(K \cdot r + \gamma_m |z| - \omega t)}$$

**Complex $\omega$**

$$E_{sp} = 2\Re e \int \frac{d^2K}{(2\pi)^2} E(K, t) \left( \mathbb{K} - \frac{K}{\gamma_m} n_m \right)$$

$$e^{i(K \cdot r + \gamma_m |z| - \omega_{sp} t)}$$

Each representation has its own dispersion relation.
General representation of the field

Which representation should be used?

Complex $K$

$$E = \int \frac{d\omega}{2\pi} \int \frac{dK_y}{2\pi} \left( \hat{K} - \frac{K_{sp}}{\gamma_m} n_m \right) E_>(K_y, \omega)$$

$$e^{i(K \cdot r + \gamma_m |z| - \omega t)}$$

The amplitude $E_>$ depends on $x$ in the sources. It does not depend on $x$ outside the sources.

The complex $k$ representation is well suited for localized stationary sources. The dispersion relation has a backbending.

General representation of the field

Which representation should be used?

Complex $\omega$

$$E_{sp} = 2\Re \int \frac{d^2K}{(2\pi)^2} E(K, t)(\hat{K} - \frac{K}{\gamma_m} n_m)$$

$$e^{i(K \cdot r + \gamma_m |z| - \omega_{sp} t)},$$

The amplitude $E_\omega$ depends on $t$ when the source is active. It does not depend on time after the sources have been turned off. The imaginary part of the frequency describes the decay of the wave.

The complex frequency formulation is well suited for pulse excitations. The dispersion relation has no backbending.

Discussion

What is the best confinement?

Localized sources and stationary regime: complex $K$ and real $\omega$

$$E = \int \frac{d\omega}{2\pi} \int \frac{dK_y}{2\pi} \left( \hat{K} - \frac{K_{sp}}{\gamma_m} \mathbf{n}_m \right) E_>(K_y, \omega) e^{i(K \cdot \mathbf{r} + \gamma_m |z| - \omega t)}$$

There is a spatial frequency cut-off for imaging applications!
Local density of states
Which choice ? Real or complex $K$?

- i) The Green’s tensor gives the answer : 
  the LDOS diverges

ii) When counting states in $k$-space, $K$ is real. We use modes with real $K$. It follows that the dispersion relation diverges.

Huygens-Fresnel principle for surface plasmons
Huygens-Fresnel principle for SP

\[ E^{SP}(x, y) = \int \frac{dk_y}{2\pi} E^{SP}(k_y) e^{i\sqrt{k_{SP}^2 - k_y^2}x + ik_yy}. \]

\[ E^{SP}(x, y) = \int dy' E^{SP}_z(x = 0, y') K(x, y, y'), \]

\[ K(x, y, y') = \begin{bmatrix} K_x \\ K_y \\ K_z \end{bmatrix} = -\frac{1}{2} \begin{bmatrix} k_z \frac{\partial^2}{\partial x^2} H^{(1)}_0(k_{SP}\rho) \\ \frac{k_z}{k_{SP}} \frac{\partial}{\partial x \partial y} H^{(1)}_0(k_{SP}\rho) \\ i \frac{\partial}{\partial x} H^{(1)}_0(k_{SP}\rho) \end{bmatrix} \]

The SPP is completely known when the z-component is known.

Teperik, Opt. Express (2009)
Asymptotic form

\[ E_z^{SP}(x, y) = \]

\[ = -\frac{i}{\sqrt{\lambda_{SP}}} \int dy' \cos \theta \ E_z^{SP}(x = 0, y') e^{ik_{SP}p} e^{i\pi/4}. \]
Huygens-Fresnel principle for SP
Huygens-Fresnel principle for SP

Influence of the number of apertures and the focal distance on the intensity at focus

![Diagram showing influence of number of apertures and focal distance on SP intensity]
Thank you!