Probing strongly coupled liquids with plasmonics

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Contents

1 Intro

You can find a very simple presentation in these lecture notes. These kind of effects have been known since the 1960s and have been studied both theoretically and experimentally.

Nowadays, have become even more relevant due to easier access to experimental techniques. An important advantage they have is due their sensitivity.

2 Surface plasmon polaritons (SPP).

This is the simplest case of a perfect interface between two different materials, e.g. air and metal, and solve Maxwell's equations:

$$
\begin{aligned}
\nabla \cdot \mathbf{D} &= \rho \\
\nabla \cdot \mathbf{B} &= 0 \\
\nabla \times \mathbf{E} &= -\partial \mathbf{B}/\partial t \\
\nabla \times \mathbf{H} &= \mathbf{J} + \partial \mathbf{D}/\partial t,
\end{aligned}
$$

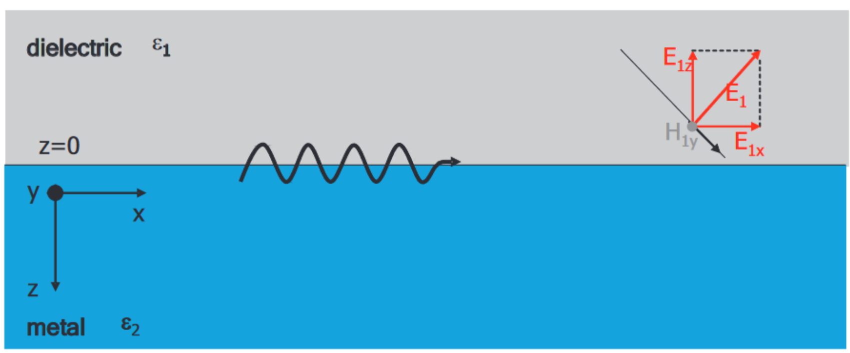
with

$$\mathbf{D} = \epsilon_0\epsilon\mathbf{E}$$
$$\mathbf{B} = \mu_0\mathbf{H}$$

Here one is making implicitly the assumption that $\epsilon = \epsilon(\omega)$, otherwise you would need to account or terms like

$$\mathbf{E}\cdot\nabla\varepsilon + \varepsilon\nabla\cdot\mathbf{E}$$

as discussed by Maier, after Eq. (2.1)



Here the crucial point is that, the dielectric function at a given frequency has to be negative, leading a mode solution for the TM (transverse magnetic) polarization that has an evanescent profile along the axis perpendicular to the plane z, whereas it can still propagate along the plane.

You start with following plane wave ansatz for each material:

$$
\begin{aligned}
\mathbf{E}_i &= (E_{i,x}, 0, E_{i,y})e^{i(\mathbf{k}_i\cdot\mathbf{r}-i\omega t)} \\
\mathbf{H}_i &= (0, H_{i,y}, 0)e^{i(\mathbf{k}\cdot\mathbf{r}-i\omega t)} \\
\mathbf{D}_i &= \epsilon_0\epsilon_i\mathbf{E}_i, \qquad \mathbf{B}_i = \mu_0\mathbf{H}_i
\end{aligned}
$$

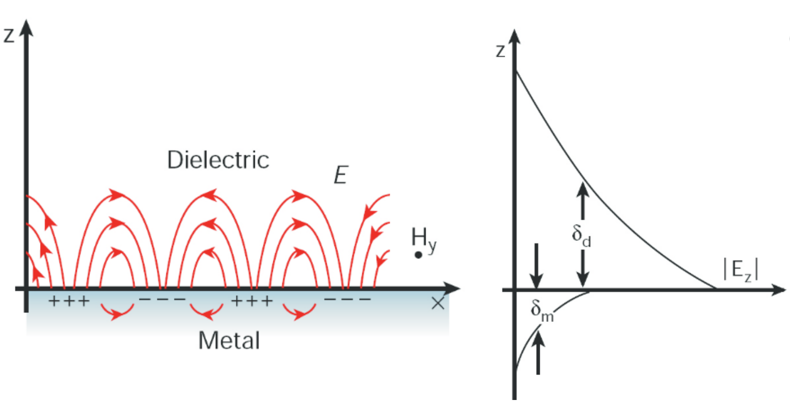
to solve Maxwell's eqs. and the following continuity relation at the air-metal interface

$$
\begin{aligned}
D_{1,z} = D_{2z}, &\qquad B_{1,z} = B_{2,z} \\
E_{1,x/y} = E_{2x/y}, &\qquad H_{1,x/y} = H_{2,x/y}
\end{aligned}
$$

This will have the relation:

$$
\frac{k_{1,z}}{\epsilon_1} = \frac{k_{2,z}}{\epsilon_2}
$$

These are indeed imaginary both. The other one left (we took $k_y = 0$ for simplicity) is the x-component k_x which is, obviously, the both materials.



And so the dispersion relation of the mode is given by:

$$k_x^2 = \frac{\omega^2}{c^2} \frac{\epsilon_1 \epsilon_2}{\epsilon_1 + \epsilon_2} \tag{1}$$

An argument of why only TM are of interest is given in Maier:

for $z < 0$. Continuity of E_y and H_x at the interface leads to the condition

$$A_1 (k_1 + k_2) = 0. \tag{2.17}$$

Since confinement to the surface requires $\mathrm{Re}[k_1] > 0$ and $\mathrm{Re}[k_2] > 0$, this condition is only fulfilled if $A_1 = 0$, so that also $A_2 = A_1 = 0$. Thus, no surface modes exist for TE polarization. *Surface plasmon polaritons only exist for TM polarization.*

This is applicable for infinite surface case, at least. One might need to review this also when dealing with a non-local dielectric function.

This is almost always solved using a Drude-like plasmon to the get:

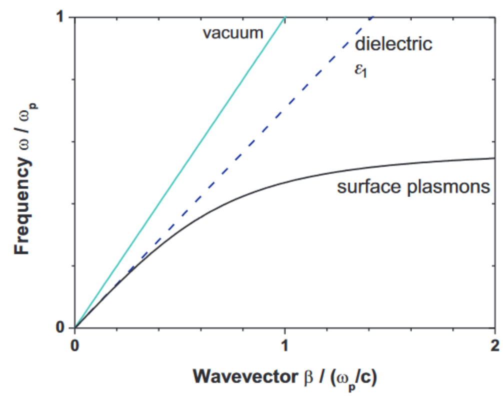
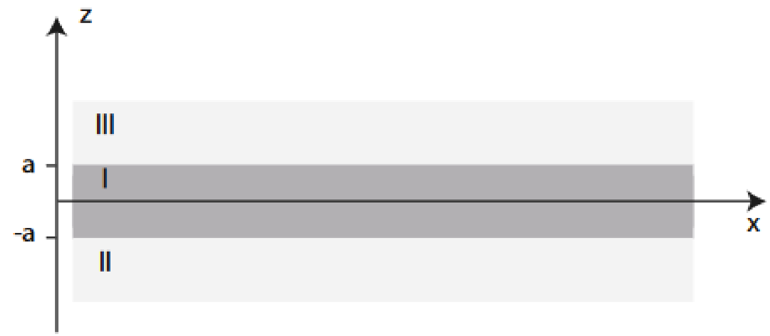


Figure 177: Dispersion relation of surface plasmons compared to light in vacuum and in the dielectric medium.

The dielectric line with respect to air or vacuum is important because SPP don't couple to any modes outside. The only way to excite them is through a prism of dielectric material whose dielectric constant is bigger than the air's one, working under total internal reflection.

3 Long-range surface plasmon polaritons

If, instead semi-infinite metal, you take a thin metal slab, sandwich between semi-infinite dielectrics:

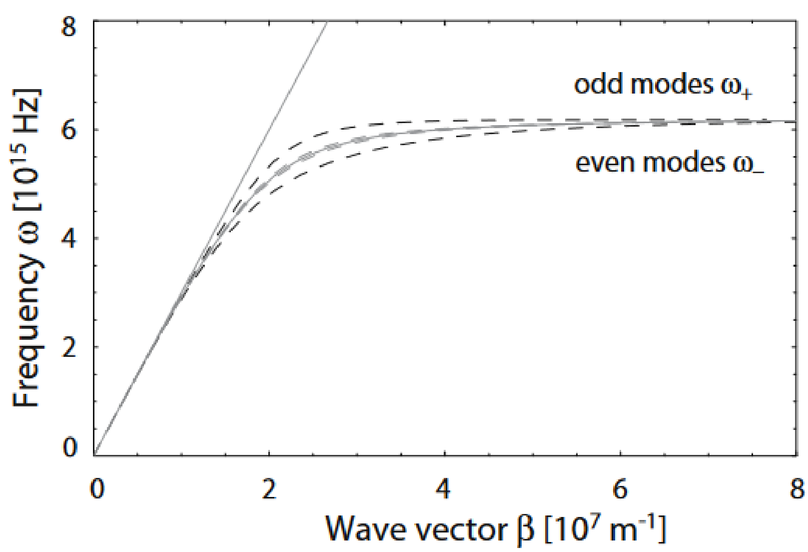


then you get two solutions that are both valid plasmon polaritons, with the following dispersion (see Plasmonics book):

$$\tanh k_1 a = -\frac{k_2 \varepsilon_1}{k_1 \varepsilon_2} \qquad (2.29a)$$

$$\tanh k_1 a = -\frac{k_1 \varepsilon_2}{k_2 \varepsilon_1}. \qquad (2.29b)$$

For a plasmon like dielectric function you get:



where the upper-mode is known as the long-range spp.

4 Surface plasmon polaritons (SPP) in 2D.

Surface plasmon polaritons also show up when considers thin metallic strips. In a recent paper on quasi-2D-metals, you can see the effect that has over the shape of the plasmon-polariton dependence on the wave-vector:

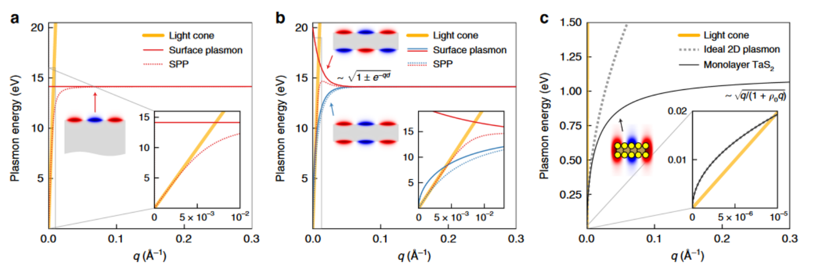


Fig. 2 Comparison of plasmon excitations on the surface of a bulk metal, on a metallic thin film, and in atomically thin metals, which are different in nature. a Dispersion relation for a surface plasmon and the lower-energy surface plasmon-polariton (SPP) branch on the surface of a three-dimensional metal with a bulk plasmon energy $\hbar\omega_p^{\mathrm{bulk}}$ ~20 eV, similar to that of bulk TaS_2. Retardation effects of the Coulomb interaction, which give rise to hybrid SPPs modes, are noticeable around the intersect of the light cone with the surface plasmon dispersion (inset). The colored intensity sketch represents the electric potential associated with the surface plasmon. **b** Same as **a**, but for a thin metallic film with thickness $d = 100$ Å. The two distinct surface plasmon modes (red and blue solid lines), as well as the two lower-energy SPP modes (red and blue dashed lines), correspond to antisymmetric and symmetric relative oscillations of the electron density on the opposite surfaces, with the symmetric mode being lower in energy. **c** Dispersion relation for monolayer TaS_2. The energy of the intrinsic plasmons in the dispersionless region is one order of magnitude smaller than that of the surface plasmons shown in **b**, and the critical wavevector $1/\rho_0 \approx 0.04$ Å^{-1} that defines the flat dispersion in monolayer TaS_2 is about an order of magnitude smaller than that of a classical, atomically thin metallic film of the same thickness, which is $1/d_{1L} = 0.2$ Å^{-1} ($\rho_0 = 25$ Å is the screening length and d_{1L} ~ 5 Å the thickness of monolayer TaS_2). Moreover, when interaction with the light field is considered, the intersection of the light cone with the intrinsic plasmon dispersion in monolayer TaS_2 occurs at a wavevector that is orders of magnitude smaller than the critical wavevector shown in Fig. 1b. Thus, the dispersionless plasmons in real quasi-2D materials that is the focus of this work are not the result of hybridizing a plasmon with a photon, i.e., it is not the same as the traditional plasmon polariton (inset).

Here, panel (a) corresponds to the semi-infinite metal, showing how the dispersion beds from a flat plasmon to the SPP. Panel (b) same but a finite-length metallic strip of 100 Angstrom thick, showing both plasmonic modes. Panel (c) corresponds to a quasi 2D metallic strip, whose thickness is about 5 Angstrom.

In the latter case, the monolayer shows the standard 2D dispersion

$$\omega_p \sim \sqrt{q} \text{ at small } q$$

Other papers for this 2D case:

- Observation of surface plasmon polaritons in 2D electron gas

- Two-dimensional Dirac plasmon-polaritons in graphene, 3D topologicalinsulator and hybrid systems

- Two-Dimensional Plasmons in Laterally Confined 2D Electron Systems

5 Papers dealing with spatial dependence for 3D semi-infinite dependence.

- Surface Plasmon in a Semi-Infinite Free-Electron Gas

- Theory of surface plasmons and surface-plasmon polaritons This is more a review than anything but treats the problem self-consistently

- Self-consistent solution of the Kohn-Sham equations for systems with inhomogeneous electron gas

- Current-induced spin polarization at the surface of metallic films: a theorem and anab initio calculation

- Conductivity of a semi-infinite electron gas: Effective "optical" surface region

- Semi-infinite jellium: Step potential model

5.1 Sum rules semi-infinite electron gas

- The Surface Dielectric Function and Its Sum Rule Semi-Infinite Electrou System

- Semi-infinite jellium: Step potential model

6 Possible problems we could work on

- The simplest case would be to deal the 3D case in the short-wavelength approximation (k=0) for metal or semiconductor, where correlations among multiple species are important. Remember that we need to have a relatively perfect interface for the polariton to propagate.

- Next would be a 2D case, where we can work out exactly all moments for arbitrary wave-number. Especially, if one can probe the case of a bilayer where there is an optic out-of-phase-mode using plasmon polaritons.

- Last, work on the full 3D case finding how the moments change with a semi-infinite metal.