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Preparatory School to the Winter College on Optics: Advances in Nano-Optics and Plasmonics

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Plasmonics

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DISPERSION

Experimentally the refractive index is a function of wavelength (frequency)

$$n(\lambda) = \sqrt{\varepsilon_r(\lambda)} \qquad\qquad \varepsilon_r(\lambda) = 1 + \chi(\lambda)$$

This phenomenon is called DISPERSION.

The polarization in a material medium can be explained considering the electrons tied to the atoms as harmonic oscillators.

Nucleus: ~2000 electron mass, i.e., infinite mass

DISPERSION

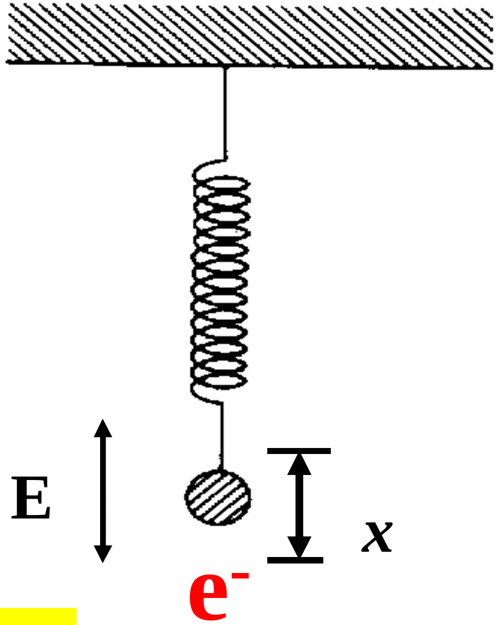
$$m\ddot{x} + \beta\dot{x} + kx = -eE_0e^{i\omega t}$$ (one-dimensional model)

Damping Restoring Force Driving Force

From the solution:

$$x = \frac{-eE_0e^{i\omega t}}{m(\omega_0^2 - \omega^2 + i\omega\gamma)} \qquad \gamma = \frac{\beta}{m}$$

$$\omega_0^2 = \frac{k}{m}$$



the induced moment is calculated:

$$p = -ex = \frac{e^2}{m\left(\omega_0^2 - \omega^2 + i\omega\gamma\right)} \cdot E_0e^{i\omega t}$$

For N oscillators per volume unit, the polarization is:

$$P = N \cdot p = \frac{N \cdot e^2}{m\left(\omega_0^2 - \omega^2 + i\omega\gamma\right)} E_0 e^{i\omega t}$$

Calling $$\alpha = \frac{\mathrm{e}^2}{\mathrm{m}(\omega_0^2 - \omega^2 + \mathrm{i}\gamma\omega)} \equiv \text{ atomic polarizability}$$

➔ $$\mathrm{P} = \mathrm{N}\alpha\mathrm{E} = \varepsilon_0 \chi \mathrm{E} \qquad \mathrm{E} = \mathrm{E}_0 \mathrm{e}^{\mathrm{i}\omega \mathrm{t}} \qquad \chi = \frac{\mathrm{N}\alpha}{\varepsilon_0}$$

where χ is the electric susceptibility.

$$\varepsilon_0 \varepsilon_\mathrm{r} = \varepsilon_0 (1 + \chi) = \varepsilon_0 \left(1 + \frac{\mathrm{N}\alpha}{\varepsilon_0}\right)$$

$$\mathrm{n}^2 = 1 + \chi = 1 + \frac{\mathrm{N}\alpha}{\varepsilon_0} \qquad \mathrm{n} = \sqrt{\varepsilon_\mathrm{r}}$$

$$n^2 = 1 + \frac{Ne^2}{\varepsilon_0 m(\omega_0^2 - \omega^2 + i\gamma\omega)}$$

If the second term is lower than 1 (as it happens in gases):

$$\boxed{n \cong 1 + \frac{Ne^2}{2\varepsilon_0 m(\omega_0^2 - \omega^2 + i\gamma\omega)}}$$

In the expression n comes out to be a complex number.

ABSTRACT

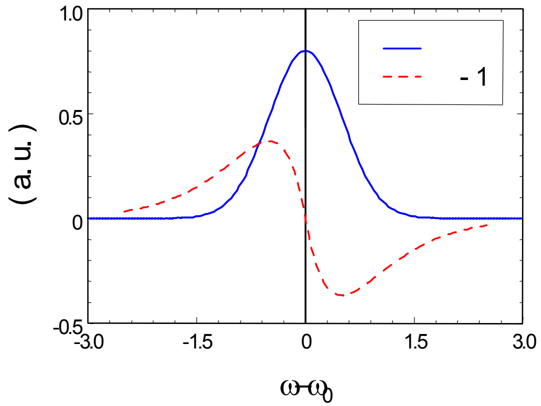
The term i$\gamma\omega$ is responsible for absorption. The complex index can be written as

$$n = \tilde{n} - i\tilde{k} = 1 + \frac{Ne^2(\omega_0^2 - \omega^2)}{2\varepsilon_0 m\left[\left(\omega_0^2 - \omega^2\right)^2 + \gamma^2\omega^2\right]} - i\frac{Ne^2\gamma\omega}{2\varepsilon_0 m\left[\left(\omega_0^2 - \omega^2\right)^2 + \gamma^2\omega^2\right]}$$

If we consider a plane wave

where $$E = A\exp\left[i(\omega t - kz)\right]$$

$$k = \omega\sqrt{\mu\varepsilon} = \frac{\omega}{c}n = \frac{2\pi}{\lambda}n$$



we see that, substituting the complex refractive index, one has

$$k = \frac{2\pi}{\lambda}(\tilde{n} - i\tilde{k})$$

which gives
$$E = A\exp\left[i\left(\omega t - \frac{2\pi\tilde{n}}{\lambda}z\right)\right]\exp\left(-\frac{2\pi\tilde{k}}{\lambda}z\right)$$

The last exponential represents a term of attenuation. The attenuation coefficient may be defined from:

$$-\alpha = \frac{1}{I}\frac{dI}{dz} \qquad I(z) = |E|^2 = I_{(0)}e^{-\alpha z}$$

By comparison with the previous equation

$$E = A\exp\left[i\left(\omega t - \frac{2\pi\tilde{n}}{\lambda}z\right)\right]\exp\left(-\frac{2\pi\tilde{k}}{\lambda}z\right)$$

$$\alpha = \frac{4\pi}{\lambda}\tilde{k}$$

METALS

In a metal the electrons are free and they do not oscillate around the atoms. **Therefore k = 0 and ω_0 = 0.**

In the equation for n^2 it is sufficient to put $\omega_0 = 0$.

$$n^2 = 1 - \frac{Ne^2}{\varepsilon_0 m(\omega^2 - i\gamma\omega)}$$

$N \equiv$ density of electrons

If $\gamma << \omega$

$$n^2 \approx 1 - \frac{\omega_p^2}{\omega^2}$$

$$\omega_p^2 = \frac{Ne^2}{\varepsilon_0 m}$$

Frequency of plasma

For Al, Cu, Au, Ag N ~ 10^{23} cm^{-3} and $\omega_P \sim 2.10^{16}$ s^{-1}.

For $\omega > \omega_P$ n is real and the waves propagate freely.

For $\omega < \omega_P$ n is pure imaginary and the field is exponentially attenuated with the distance from the surface. Therefore the radiation is reflected from the surface.

Therefore, for visible radiation and infrared ω < ω_P and n is imaginary. In general, n is complex because there is γ:

$$1-n^2=\frac{Ne^2}{\omega\varepsilon_0 m(\omega-i\gamma)}\frac{(\omega+i\gamma)}{(\omega+i\gamma)}=\frac{Ne^2}{\omega\varepsilon_0 m\left(\omega^2+\gamma^2\right)}+i\gamma\frac{Ne^2}{\omega\varepsilon_0 m\left(\omega^2+\gamma^2\right)}=$$

$$\frac{\omega_p}{\left(\omega^2+\gamma^2\right)}+i\frac{\omega_p}{\left(\omega^2+\gamma^2\right)}\frac{\gamma}{\omega}$$

From the refractive index expression we may derive the dielectric function

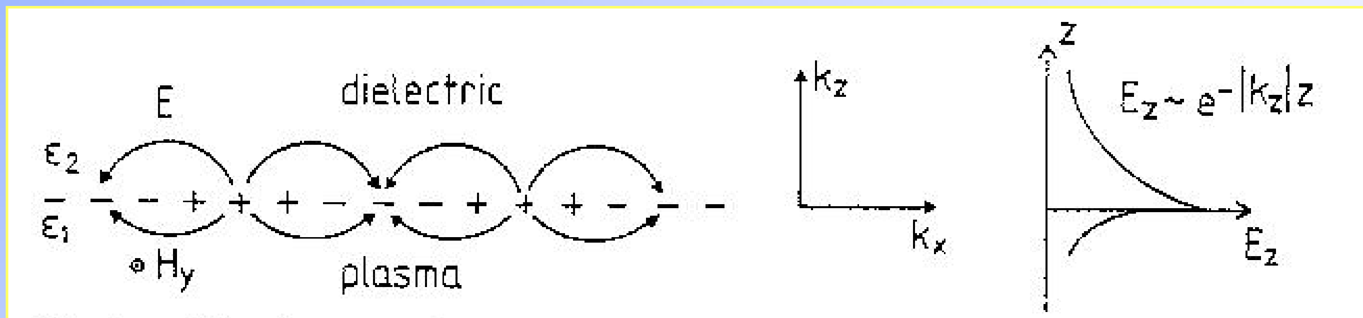
$$\varepsilon \simeq 1 - (\omega_p / \omega)^2$$

This is the so-called Drude expression for the dielectric function in a metal-

It is positive for $\omega > \omega_p$ and negative for $\omega < \omega_p$

SURFACE PLASMONS ON SMOOTH SURFACES

The electron charges on a metal boundary can perform coherent fluctuations which are called surface plasma oscillations. The frequency ω of these longitudinal oscillations is tied to its wave vector k_x by a dispersion relation $\omega(k_x)$.



These charge fluctuations, which can be localized in the z direction within the Thomas-Fermi screening length of about 1 Å, are accompanied by a mixed transversal and longitudinal electromagnetic field which disappears at $|z| \to \infty$ (fig.1) and has its maximum in the surface z = 0, typical for surface waves. This explains their sensitivity to surface properties.

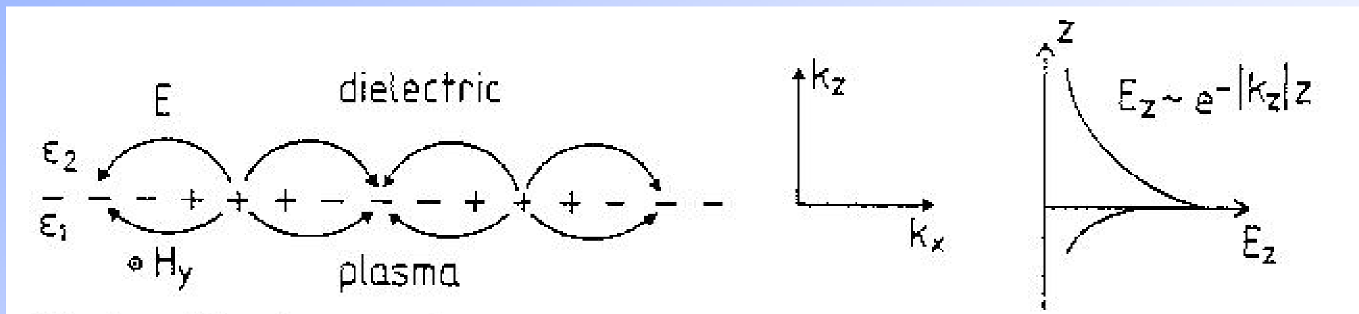


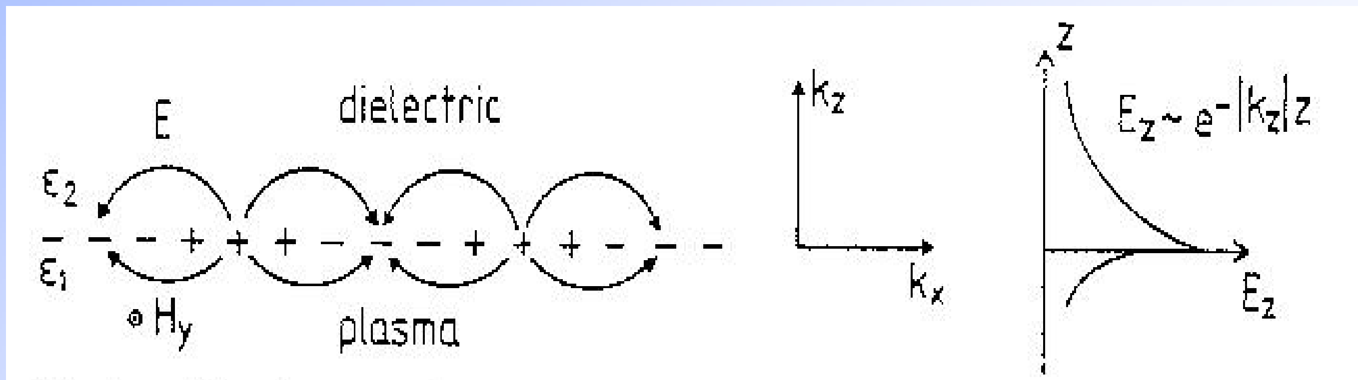
Fig.1 The charges and the electromagnetic field of SPs propagating on a surface in the x direction are shown schematically. The exponential dependence of the field E_z is seen on the right. H_y shows the magnetic field in the y direction of this p-polarized wave.

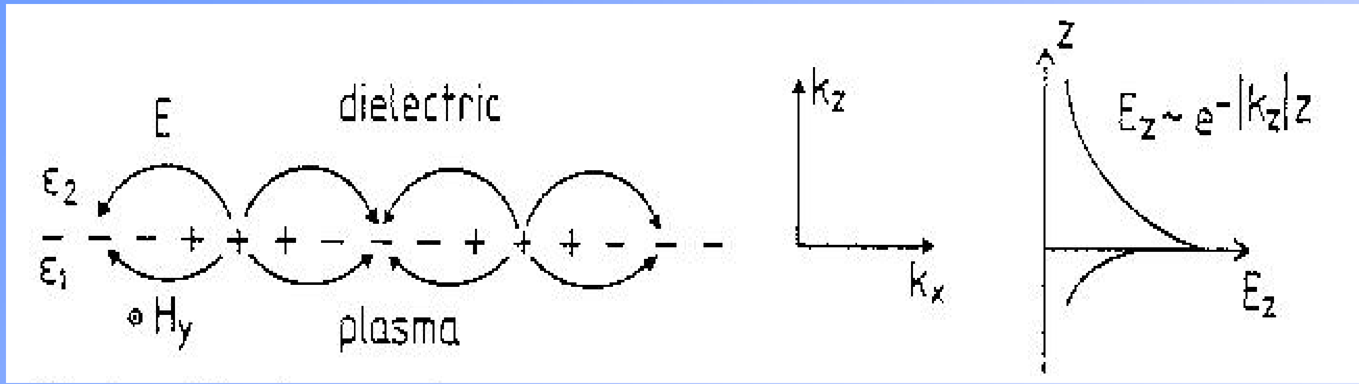
The field is described by

$$E = E_0^{\pm} \exp\left[+i\left(k_x x \pm k_z z - \omega t\right)\right] \quad (1)$$

with + for z≥0, - for z≤0, and with imaginary k_z, which causes the exponential decay of the field E_z.

The wave vector k_x lies parallel to the x direction; k_x = $2\pi/\lambda_p$, where λ_p is the wavelength of the plasma oscillation.





The wave propagates in the x-direction. The problem does not depend on y. The field is

$$z>0 \qquad H_2=(0,H_{2y},0)\exp i\left(k_{2x}x+k_{2z}z-\omega t\right)$$

$$E_2=\left(E_{2x},0,E_{2z}\right)\exp i\left(k_{2x}x+k_{2z}z-\omega t\right) \qquad (1)$$

$$z<0 \qquad H_1=(0,H_{1y},0)\exp i\left(k_{1x}x-k_{1z}z-\omega t\right)$$

$$E_1=\left(E_{1x},0,E_{1z}\right)\exp i\left(k_{1x}x-k_{1z}z-\omega t\right) \qquad (2)$$

The fields obey to Maxwell equations

$$rot\, H_i = \mu_i \varepsilon_i \frac{\partial E_i}{\partial t} \;\therefore\; rot\, H_i = \varepsilon_i \frac{\partial E_i}{\partial t} \qquad (3)$$

$$rot\, E_i = -\frac{\partial B_i}{\partial t} \qquad \therefore\; rot\, E_i = -\mu \frac{\partial H_i}{\partial t} \qquad (4)$$

$$div\, \varepsilon_i E_i = 0 \qquad (5)$$

$$div\, H_i = 0 \qquad (6)$$

with the continuity conditions

$$\boxed{B = \mu H = \frac{E}{c}}$$

$$E_{1x} = E_{2x} \qquad (7)$$

$$H_{1y} = H_{2y} \quad \rightarrow \quad \frac{E_{1z}}{v_1 \mu} = \frac{E_{2z}}{\mu v_2} \qquad (8)$$

$$\sqrt{\varepsilon_1} E_{1z} = \sqrt{\varepsilon_2} E_{2z} \qquad (9)$$

$$\boxed{\mu v_1 = \frac{\mu}{\sqrt{\varepsilon \mu}} = \sqrt{\frac{\mu}{\varepsilon}}}$$

Taking (1) and (2) at z = 0

$$
\begin{aligned}
z>0 \qquad & H_2=(0,H_{2y},0)\exp i\left(k_{2x}x+k_{2z}z-\omega t\right) \\
& E_2=\left(E_{2x},0,E_{2z}\right)\exp i\left(k_{2x}x+k_{2z}z-\omega t\right) \qquad (1) \\
z<0 \qquad & H_1=(0,H_{1y},0)\exp i\left(k_{1x}x-k_{1z}z-\omega t\right) \\
& E_1=\left(E_{1x},0,E_{1z}\right)\exp i\left(k_{1x}x-k_{1z}z-\omega t\right) \qquad (2)
\end{aligned}
$$

$$
\begin{aligned}
H_2 &= H_{2y}\exp i\left\{k_{2x}x-\omega t\right\} && (10a) \\
E_2 &= E_{2x}\exp i\left\{k_{2x}x-\omega t\right\} && (10b) \\
&= E_{2z}\exp i\left\{k_{2x}x-\omega t\right\} && (10c) \\
H_1 &= H_{1y}\exp i\left\{k_{1x}x-\omega t\right\} && (10d) \\
E_1 &= E_{1x}\exp i\left\{k_{1x}x-\omega t\right\} && (10e) \\
&= E_{2z}\exp i\left\{k_{2x}x-\omega t\right\} && (10f)
\end{aligned}
$$

Because $E_{1x} = E_{2x}$ it follows

$$E_{1x}\exp i(k_1x-\omega t)=E_{2x}\exp i\left\{k_2x-\omega t\right\}$$

therefore

$$k=k_{1x}=k_{2x} \qquad (11)$$

From $rot\, H = \varepsilon \frac{\partial E}{\partial t}$

$$rot\, H_i = \begin{vmatrix} \vec{i} & \vec{1} & \vec{k} \\ \frac{\partial}{\partial x} & \frac{\partial}{\partial y} & \frac{\partial}{\partial z} \\ H_x & H_y & H_z \end{vmatrix} = -\vec{i}\,\frac{\partial H_{yi}}{\partial z} = \varepsilon_i \frac{\partial E_{ix}}{\partial t}\,\vec{i}$$

or

$$\frac{\partial H_{yi}}{\partial z} = \omega \varepsilon_i E_{1x}$$

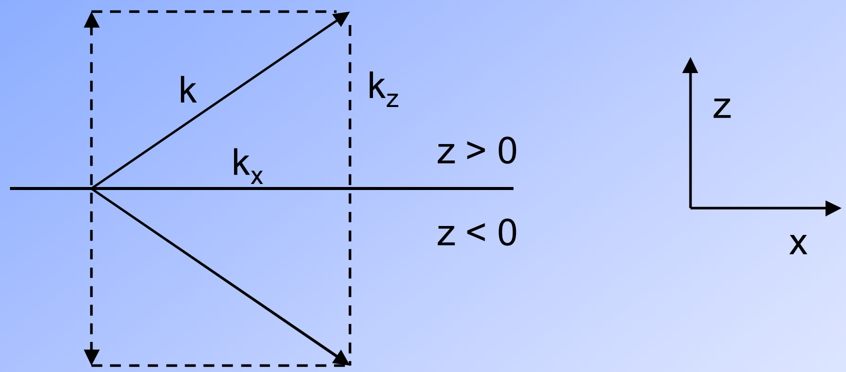
$$\begin{array}{lll} z > 0 & H_2 = (0, H_{2y}, 0) \exp i\left(k_{2x} x + k_{2z} z - \omega t\right) & \\ & E_2 = \left(E_{2x}, 0, E_{2z}\right) \exp i\left(k_{2x} x + k_{2z} z - \omega t\right) & (1) \\ z < 0 & H_1 = (0, H_{1y}, 0) \exp i\left(k_{1x} x - k_{1z} z - \omega t\right) & \\ & E_1 = \left(E_{1x}, 0, E_{1z}\right) \exp i\left(k_{1x} x - k_{1z} z - \omega t\right) & (2) \end{array}$$

that is

$$\begin{aligned} k_{2z} H_{y2} &= \omega \varepsilon_2 E_{x2} \\ -k_{1z} H_{y1} &= \omega \varepsilon_1 E_{x1} \qquad (12) \end{aligned}$$

Moreover

$$k_x^2 + k_{z1}^2 = \varepsilon_i \left(\frac{\omega}{c}\right)^2 \qquad (16)$$



or

$$k_x^2 = \varepsilon_i \left(\frac{\omega}{c}\right)^2 - k_{zi}^2$$

Because $E_{x2} = E_{x1}$ it is

$$\frac{k_{2z}}{\varepsilon_2} H_{2y} = \omega E_{x2} \quad \text{e} \quad \frac{k_{1z}}{\varepsilon_1} H_{1y} = -\omega E_{x1} \qquad (13)$$

summing ($E_{x2} = E_{x1}$)

$$\frac{k_{1z} H_{1y}}{\varepsilon_1} + \frac{k_{2z} H_{2y}}{\varepsilon_2} = 0 \qquad (14)$$

But because $H_{1y} = H_{2y}$ it is

$$k_{2z} / \varepsilon_2 + k_{1z} / \varepsilon_1 = 0 \qquad (15)$$

Finally we have

$$K^2_x = \varepsilon_1(\omega/c)^2 - k^2_{z1}$$

$$K^2_x = \varepsilon_2(\omega/c)^2 - k^2_{z2}$$

$$K_{1z} / \varepsilon_1 = -K_{2z} / \varepsilon_2$$

$$k_x^2 = \varepsilon_1 \left(\frac{\omega}{c}\right)^2 - k_{z1}^2 \qquad \frac{k_{1z}}{\varepsilon_1} = -\frac{k_{2z}}{\varepsilon_2}$$

$$k_x^2 = \varepsilon_2 \left(\frac{\omega}{c}\right)^2 - k_{z2}^2$$

$$2k_x^2 = \left(\frac{\omega}{c}\right)^2 (\varepsilon_1 + \varepsilon_2) - k_{z1}^2 - \frac{k_{1z}^2 \varepsilon_2^2}{\varepsilon_1^2}$$

$$= \left(\frac{\omega}{c}\right)^2 (\varepsilon_1 + \varepsilon_2) - k_{z1}^2 \left(1 + \frac{\varepsilon_2^2}{\varepsilon_1^2}\right)$$

$$= \left(\frac{\omega}{c}\right)^2 (\varepsilon_1 + \varepsilon_2) - \left[k_x^2 + \varepsilon_1 \left(\frac{\omega}{c}\right)^2\right] \left(1 + \frac{\varepsilon_2^2}{\varepsilon_1^2}\right)$$

$$k_x^2 \left(1 - \frac{\varepsilon_2^2}{\varepsilon_1^2}\right) = \left(\frac{\omega}{c}\right)^2 (\varepsilon_1 + \varepsilon_2) - \left(\frac{\omega}{c}\right)^2 \varepsilon_1 \left(1 + \frac{\varepsilon_2^2}{\varepsilon_1^2}\right)$$

$$k_x^2\frac{\varepsilon_1^2-\varepsilon_2^2}{\varepsilon_1^2}=\left(\frac{\omega}{c}\right)^2\left[\varepsilon_1+\varepsilon_2-\varepsilon_1-\frac{\varepsilon_2^2}{\varepsilon_1}\right]$$

$$=\left(\frac{\omega}{c}\right)^2\varepsilon_2\left(1-\frac{\varepsilon_2}{\varepsilon_1}\right)=\left(\frac{\omega}{c}\right)^2\varepsilon_2\frac{\varepsilon_1-\varepsilon_2}{\varepsilon_1}$$

$$k_x^2\frac{(\varepsilon_1-\varepsilon_2)(\varepsilon_1+\varepsilon_2)}{\varepsilon_1}=\left(\frac{\omega}{c}\right)^2\varepsilon_2(\varepsilon_1-\varepsilon_2)$$

$$k_x^2=\left(\frac{\omega}{c}\right)^2\left(\frac{\varepsilon_2\varepsilon_1}{(\varepsilon_1+\varepsilon_2)}\right)$$

$$k_x=\frac{\omega}{c}\left(\frac{\varepsilon_1\varepsilon_2}{(\varepsilon_1+\varepsilon_2)}\right)^{1/2} \qquad (17)$$

If we assume $\varepsilon_2 = 1$ (air) and $\varepsilon_1 < 0$ (metal) with $|\varepsilon_1| > \varepsilon_2$

k_x is real and

$$k_{zi}^2 = \varepsilon_i \left(\frac{\omega}{c}\right)^2 - k_x^2$$

Because in this case $k_x > \dfrac{\omega}{c}$

$$k_{2z}^2 = \left(\frac{\omega}{c}\right)^2 - k_x^2 < 0$$

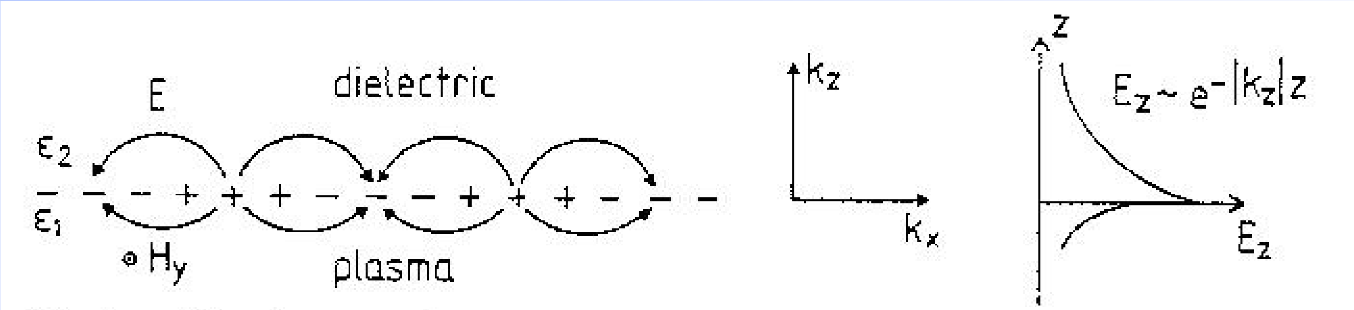
and k_{2z} and k_{1z} are immaginary or complex.

Maxwell's equations yield the retarded dispersion relation for the plane surface of a semi-infinite metal with the dielectric function $(\varepsilon_1 = \varepsilon_1' + i\varepsilon_1''),$ adjacent to a medium ε_2 as air or vacuum:

$$D_0 = \frac{k_{z1}}{\varepsilon_1} + \frac{k_{z2}}{\varepsilon_2} = 0 \quad \text{together with} \qquad (2)$$

$$\varepsilon_1 \left(\frac{\omega}{c}\right)^2 = k_x^2 + k_{zi}^2 \quad \text{or} \qquad (3)$$

$$k_{zi} = \left[\varepsilon_i \left(\frac{\omega}{c}\right)^2 - k_z^2\right]^{1/2}, \quad i = 1, 2.$$



The wave vector k_x is continuous through the interface. The dispersion relation can be written as

$$k_x = \frac{\omega}{c}\left(\frac{\varepsilon_1 \varepsilon_2}{\varepsilon_1 + \varepsilon_2}\right)^{1/2}. \qquad (4)$$

If we assume, besides a real ω and ε_2 ,a complex ε_1 with $\varepsilon_1'' < |\varepsilon_1'|$, we obtain a complex $k_x = k_x' + i k_x''$ with

$$k_x' = \frac{\omega}{c}\left(\frac{\varepsilon_1' \varepsilon_2}{\varepsilon_1' + \varepsilon_2}\right)^{1/2} \qquad (5)$$

$$k_x'' = \frac{\omega}{c}\left(\frac{\varepsilon_1' \varepsilon_2}{\varepsilon_1' + \varepsilon_2}\right)^{3/2} \frac{\varepsilon_1''}{2(\varepsilon_1')^2}. \qquad (6)$$

For real k'_x one needs $\varepsilon'_1 < 0$ and $|\varepsilon_1'| > \varepsilon_2,$ which can be fulfilled in a metal and also in a doped semiconductor near the eigen frequency; k_2'' determines the internal absorption. In the following we write k_x in general instead of k'_x.

Let us take ε_2 real and $\varepsilon_1 = \varepsilon'_1 + i\varepsilon''_1$

$$k_x^2 = \left(\frac{\omega}{c}\right)^2 \frac{\left(\varepsilon_1' + i\varepsilon_1''\right)\varepsilon_2}{\varepsilon_1' + i\varepsilon_1'' + \varepsilon_2} = \left(k_x' + ik_x''\right)^2 = k_x'^2 - k_x''^2 + 2ik_x'k_x''$$

$$= \left(\frac{\omega}{c}\right)^2 \frac{\left(\varepsilon_1' + i\varepsilon_1''\right)\varepsilon_2\left[\left(\varepsilon_1' + \varepsilon_2\right) - i\varepsilon_1''\right]}{\left(\varepsilon_1' + \varepsilon_2\right)^2 + \varepsilon_1''^2}$$

$$k_x'^2 - k_x''^2 = \left(\frac{\omega}{c}\right)^2 \frac{\varepsilon_2}{\left(\varepsilon_1' + \varepsilon_2\right)^2 + \varepsilon_1''^2}\left[\varepsilon_1'\left(\varepsilon_1' + \varepsilon_2\right) + \varepsilon_1''^2\right]$$

$$2k_x'k_x'' = \left(\frac{\omega}{c}\right)^2 \frac{\varepsilon_2\left[-\varepsilon_1'\varepsilon_1'' + \varepsilon_1''\left(\varepsilon_1' + \varepsilon_2\right)\right]}{\left(\varepsilon_1' + \varepsilon_2\right)^2 \varepsilon_1''^2}$$

From which

$$k_x' = \frac{\omega}{c}\left(\frac{\varepsilon_1'\varepsilon_2}{\varepsilon_1' + \varepsilon_2}\right)^{1/2}$$

$$k_x'' = \frac{\omega}{c}\left(\frac{\varepsilon_1'\varepsilon_2}{\varepsilon_1' + \varepsilon_2}\right)^{3/2}$$

The dispersion relation (see figure) approaches the light line $\sqrt{\varepsilon_2}\omega/c$ at small k_x, but remains larger than $\sqrt{\varepsilon_2}\omega/c$ so that the SPs cannot transform into light: it is a “nonradiative” SP.

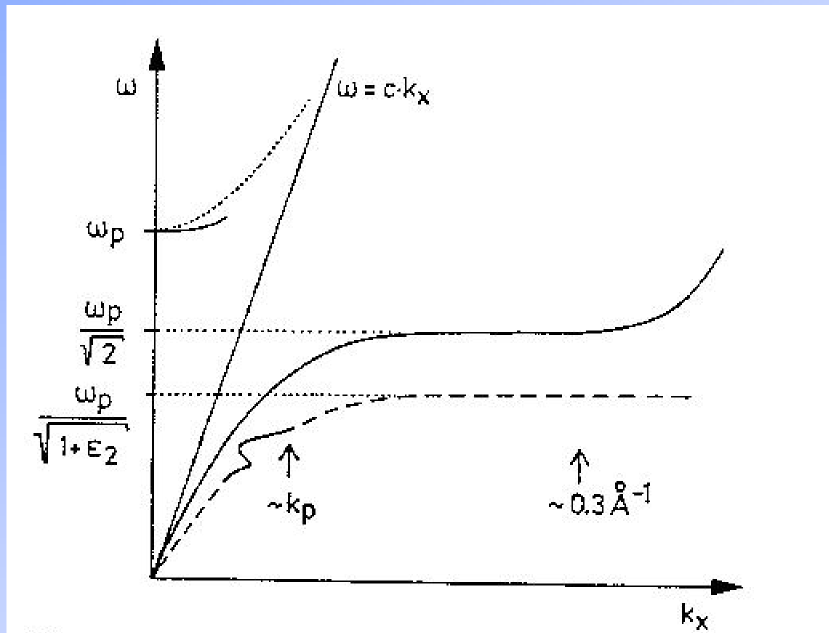


Fig.2. The dispersion relation of nonradiative SPs (—), right of the light line $\omega = ck_x$; the retardation region extends from $k_x = 0$ up to about $k_p = 2\pi/\lambda_p$ (λ_p plasma wavelength). The dashed line, right of $\omega = ck_x$, represents SPs on a metal surface coated with a dielectric film (ε_2). Left of the light line, $\omega(k_x)$ of the radiative SPs starts at ω_p (—). The slight modulation in the dashed dispersion curve comes from an eigen frequency in a monomolecular dye dilm deposited on a Langmuir-Blodgett film (ε_2).

By substituting the expression of the dielectric constant of metals k_x may be written as

$$k_x = (\omega//c)\sqrt{\ }\{(\omega^2 - \omega_p^2)/(2\omega^2 - \omega_p^2)\}$$

When k is very large it should be

$$\omega = \omega_p \sqrt{2}$$

And more in general if the first medium is not air

$$\omega_{sp} = \left[\frac{\omega_p}{1+\varepsilon_2}\right]^{1/2} \qquad (8)$$

At large k_x or

$$\varepsilon_1' \rightarrow -\varepsilon_2 \qquad (7)$$

the value of ω approaches

$$\omega_{sp} = \left[\frac{\omega_p}{1+\varepsilon_2}\right]^{1/2} \qquad (8)$$

for a free electron gas where ω_p is the plasma frequency $\sqrt{4\pi ne^2/m}$, with n the bulk electron density.

With increasing ε_2, the value of ω_{sp} is reduced.

At large k_x the group velocity goes to zero as well as the phase velocity, so that the SP resembles a localized fluctuation of the electron plasma.