International Journal of Scientific Engineering and Research (IJSER)
ISSN (Online): 2347-3878
Index Copernicus Value (2015): 62.86 | Impact Factor (2015): 3.791

Surface Plasmon's in Non Linear Response Function in Metal Optics

T M Ehteshamul Haque

Abstract: ***If one interpret the surface layer as the selvedge region of the metal half space, differing from thebulk metal only by a reduced density of free electrons, we deal with the hydrodynamic model used to discuss surface electro magnetic fields. The "local Plasmon" excitation discussed in that context is physically the same thing as a standing plasma wave in the surface layer. Surface resonances of this type are closely related to the "multiple surface Plasmon's"***

Keywords: Surface Plasmon's, Feibeleman's Treatment, Surface Plasmon's mode, Higher multiple mode, Quantum mechanical RPA calculations of MSP.

1. Introduction

In the preceding discussion, excitation of standing plasma waves in the surface layer was related to pole type singularities of $d_{\perp}$ (ω) -$d_{||}$(ω) or, equivalently, of $n_z^{(1)}(0)$ given by (5.31). It must understood that a genuine singularity of $n_z^{(1)}(0)$ as a function of ω can occur only in the LWL. Such a singularity results from vanishing denominator of the LWL of λ.

The ratio E_x^1/E_x^t of the x component of longitudinal and transverse electric field being usually of the order of $k_x^2/P_tP_1 \ll 1$, is now of order unity, since a strong longitudinal field is accompanied with the standing wave. As a consequence, the value of $n_z^{(1)}(0)$given by (5.22) is, at the renance frequency, enhanced by a factor of P_tP_1/k_x^2. In the LWL this enhancement appears as a singularity.

2. Surface Plasmon's

One generalized the treatment of surface Plasmon's in two directions. First, our express the Plasmon dispersion relation in terms of the surface response functions $d_{\perp}$ (ω) -$d_{||}$(ω) and obtain a general result which applies not only to the hydrodynamic model but also to microscopic surface models. In this part one follow FELIBELMAN'S [1-3]. Second one extend the discussion to the so called "multipole surface plasmons"(MSP), which are additional surface modes including standing plasma waves in a surface region of low electron density. The possible existence of such modes at clean metal surfaces has been a matter of controversy for more than a decade[10-13]. Our discussion, following recent work of KEMPA and GERHARDTS[14] will establish a relation between the MSP and the photoemission experiment on aluminium first pointed out by SCHWARTZ and SCHAICH[15]. The explicit results of sect. is helpful for a qualitative understanding of the frequency dependence of $d_{\perp}$. The LWL is taken throughout the whole section.

3. Feibeleman's Treatment

A surface Plasmon at a metal / vacuum interface can be defined as an Eigen solution of Maxwell's equations with an electric field propagation along the interface and decaying exponentially into both the metal and the vacuum. Such an eigemode exists if the reflection amplitude for p-polarized light becomes singular for

$$P_a = iP_a, \quad P_t = iP_t, \tag{5.36}$$

And $P_a > 0$, $P_t > 0$. Then finite reflected and transmitted fields, both decaying way from the surface, are possible for vanishing incident field. Inserting (5.36) into (5.8), one obtain as condition for the existence of a surface Plasmon that the denominator of

$$r_p = \frac{\varepsilon_t p_a - \varepsilon_a p_t - i(\varepsilon_a - \varepsilon_t)(p_a p_t d_\pi - k_x^2 d_\perp)}{\varepsilon_t p_a + \varepsilon_a p_t - i(\varepsilon_a - \varepsilon_t)(p_a p_t d_\pi - k_x^2 d_\perp)} \tag{5.37}$$

must vanish. From the dispersion of transverse waves.

$$k_x^2 + p_a^2 = \varepsilon_a \omega^2 / c^2 = P_t^2 + \varepsilon_t \omega^2 / c^2 \tag{5.38}$$

One has

$$(\varepsilon_t p_a + \varepsilon_a P_t)(p_a - P_t) = (\varepsilon_a - \varepsilon_t)(p_a P_t - k_x^2) \tag{5.39}$$

If one assumes $|P_a d_\Pi| \sim |k_x d| < \frac{1}{2}$, then (5.39) show that for vanishing denominator of (5.37) the difference P_aP_t and k_x^2. Hence, in the LWL, the condition for a surface Plasmon can be written as

$$\varepsilon_t p_a + \varepsilon_a P_t + (\varepsilon_t - \varepsilon_a)\left(d_\perp - d_\Pi\right) \text{Pa Pt} = 0 \tag{5.40}$$

This form of the surface Plasmon dispersion relation (SPDR) has been derived by FEIBELMAN1[-3] and in similar form (but with less transparent methods) also by other authors[16-18]. Since (5.40) depends only on the difference $d_\perp - d_\Pi$, the SPDR is not changed if the surface position is shifted by an amount a, whereas the values of both $d_\perp$andd_Π are changed by that amount a, as is easily seen from. This consistency requirement is also satisfied by the form

$$\varepsilon_t p_a + \varepsilon_a P_t + (\varepsilon_t - \varepsilon_a)\left(d_\perp - d_\Pi\right) k_x^2 = 0 \tag{5.41}$$

Which is completely equivalent to (5.40) reduces fro the HD with a single step electron density profile ($d_\Pi = 0, d_\perp = /P_1$with $\varepsilon_a = \varepsilon_b = 1$) exactly to the SPDR with notation $\lambda_0 =, \lambda - iP_{t,} \varepsilon = \varepsilon_t, n = P_1$) which is not restricted to the long wavelength limit.

In the non-retarted($c \to \infty$)limit, (5.38) yields P=P_a=K_x, and both (5.70) (5.41) reduce to

$$\varepsilon_t + \varepsilon_a + (\varepsilon_t - \varepsilon_a)\left(d_\perp - d_\Pi\right) K_x = 0 \tag{5.42}$$

With the free electron dielectric constant, $\varepsilon_t = 1 - \omega_p^2/\omega^2$, this takes for small K_x, the form

$$\omega_s = \frac{\omega_p}{\sqrt{\varepsilon_a+1}}\left[1 + \frac{\varepsilon_a}{\varepsilon_a+1}(d_\perp - d_\Pi)K_x\right] \quad (5.43)$$

For the SPDP, where $d_\perp$-d_Π is taken $\omega = \omega_s \approx \omega_p/(\varepsilon_a + 1)^{1/2}$. For this surface Plasmon the induced charge density $p^{ind}(z)$ is dominated by a single peak in the surface region, since $4\pi p^{ind}(z) = V.E. \approx dE/dz$, and $E_z(z)$ interpolates smoothly between the nearly constant (in the LWL) classical values outside and inside the metal. Then according to $d_\perp$ (ω) measures the position of this peak i.e. the mean position of the induced charge. Sign and value of $d_\perp$-d_Π depend crucially on the diffuseness of the surface. If the electron density of the unperturbed metal is assumed to drop at the surface abruptly from the constant bulk value to zero, as in the simple hydrodynamic model or in the "semi classical infinite barrier" model, but also if the model density profile is too steep, as for the microscopic infinite barrier model, the induced charge lies on the metal side of the surface (of the jellim edge for the IBM) and $d_\perp$-$d_\Pi$comes out positive. For the soft, sel-consistent Lang_Kohn profile, on the other hand, the induced charge is located essentially outside the jellim edge and $d_\perp$-$d_\Pi$negative. Within the hydrodynamic model this can be simulated by a suitably choose surface layer of reduced electron density. If retardation effects are included, the negative values of $d_\perp$-$d_\Pi$obtained for diffuse surfaces lead to a plateau in the surface-plasmon dispersion relation.

4. Additional Surface Plasmon Modes

In order to understand how additional surface plasmons can be discussed in terms of the surface response function $d_\perp$ (ω) -$d_{||}$(ω), it is instructive to assume that $d_\perp$ (ω) exhibits at a certain "resonance frequency" ω_r a pole singularity, e.g.,

$$d_\perp\text{-}d_\Pi \approx \frac{a_r\omega_r}{\omega-\omega_r} \; for \; \omega \approx \omega_r \quad (5.44)$$

One has discussed that this can happen for instance, in the nonlocal three layer model if the frequency is larger than the plasma frequency of the surface layer, but less than the plasma frequency of the bulk metal. Then, in addition to the "regular" surface plasmon, (5.43), the dispersin relation (5.42) yields a branch with frequency near ω_r (for small) positive values of k_x),

$$\omega = \omega_r\left[1 + \frac{\varepsilon_a-\varepsilon_t}{\varepsilon_a+\varepsilon_t} a_r k_x\right] \quad (5.45)$$

where the bulk dielectric function $\varepsilon_t = 1 - (\omega_p/\omega)^2$ is taken at $\omega = \omega_r$. These additional surface Plasmon modes are accompanied with standing plasma waves in the surface region of reduced electron density, BENNETT[19] first pointed at their existence, and EQUILUZ et at[20-23], who discussed their appearance for arbitrarily shaped density profile, addressed them as "higher multipole" modes, since in the non retarded limit the total induced charge of these additional modes was found to vanish. If retardation effects are properly taken into account, this is no longer true. BOARDMAN et al[24] investigated in detail the electric field and the fluctuation charge density in this case and obtained an oscillatory behaviour of the latter, although not a clear multipole structure.

Quantum mechanical RPA calculations of MSP modes in the non retarded limit have been presented by INGLESFIELD and WIKBORD25, who used a double step function to simulate the effective potential (not the density) of conduction electrons at a n aluminium surface covered with an over layer of alkali atoms. "Multipole" modes were found for over layers with a sufficiently extended how density region, but not for a single step potential, which was considered as a reasonable model of uncoated Al.

Having related the existence of a MSP mode to a pole singularity of $d_\perp(\omega)$, we should understand how such a singularity can be consistent with telling that $d_\perp(\omega)$ is the "center of gravity"of the fluctuation charge density. Since $d_\perp(\omega)$ is independent of the wave number k_x, we can calculate $d_\perp(\omega)$, from the fields excited by an external plane wave impinging on the surface rather than by the fields related to a surface Eigen mode with $k_x > \omega/c$. According to the total fluctuation charge, determined by the transverse fields far from the surface, is insensitive to details of the surface region. Especially, it can not vanish at the resonance frequency ω_r for excitation of a standing plasma wave in the low density surface region in order to produce the singularity of $d_\perp(\omega)$. On the other hand, near ω_r the induced charge density will exhibit a spatial oscillation in the surface region and the numerator of measuring the dipole moment of this charge distribution, will diverge at resonance, $\omega = \omega_r$, since then the amplitude of this spatial oscillation is enhanced by a factor ($\sim P_t P_\ell / k_x^2$ is discussed at the end of Sect.) which diverges in the LWL. Moreover, as the frequency seeps through the resonance, the phase of the excited plasma wave in the surface layer will change, so that the induced dipole moment changes sign out ω_r, whereas the total induced charge is completely insensitive to these surface effects. Thus, we see that the resonant excitation of standing plasma waves in the low density surface region indeed leads, in the LWL, to a pole structure 0, $d_\perp(\omega)$. Near the pole, $d_\perp(\omega)$. should be interpreted as dipole moment rather than as mean position of the induced charge distribution.

So far discussion of MSP modes has neglected damping effects. If damping is included, the pole structure of $d_\perp(\omega)$. issmeared out, its imaginary part becomes a broadened δfunction peaked at ω_r and its real part exhibits the S-like shape of a smeared out principle value function. The reflection amplitude r_p (5.37) no longer diverge for real values of ω and k_x, and surface Plasmon's are damped. Nevertheless, for sufficiently small damping, a damped Eigen mode is expected to

5. Discussion of Result

In this chapter, we have evaluated inverse reflectivity as a function of ω/ω_p. We have used the value of surface response function $d_\perp$ calculated from Chapter IV for different values of k_x/k_F. We have taken the value of k_x/k_F as 0.005, 0.05 and 0.1 respectively. There evaluation has been performed from Feibelman's treatment. Relection coefficient in the surface regime ($k_x > \omega/c$) has been calculated from equation (5.37) with Feibelman's $d_\perp(\omega)$

result for r_s=4. Feibelman's $d_\perp(\omega)$ result for r_s=4 IS RPA result. From our result it indicates that with increasing value of $K_x 1/|r_p|^2$ increases very fastly. We have also evaluated $1/|r_p|^2$ with hydrodynamic approximation keeping the value of k_x/k_F = 0.1 and surface damping $r_s=0.3\omega_p$ and $0.15\omega_p$. In there calculation $1/|r_p|^2$ gives a peak and after that it decay. The peak is must more proved the value of $r_s=0.1\omega_p$. These peaks are supposed to be damped multipole surface Plasmon.

This result is not really in conflict with the calculation of Inglishfield and Kborg[36] who used a single-stop model for the electron potential at a clean surface. The resulting density profile is similar to that for the IBM and much deeper than the self-consistent lang-kohn profile. RPA calculation for the IBM[37] also yield no MSP mode for r_s values in the metallic range (2 to 6)[38]. Since the surface region of low density is too small. This region increases with increasing values of r_s and for very large r_s values (for bulk densities) MSP modes appear in the quantum mechanical mode. Recent microscopic calculation for charge surface support their arguements[39].

Finally one wants to emphasise an important difference between hydrodynamic and RPA calculations, namely the role played by damping effects. In hydrodynamic calculation damping (r,r_s) appears as a free parameter which can be neglected completely $(r=r_s=0)$ for multipole surface Plasmon. In RPA calculation damping mechanism is included automatically, namely optical excitation of electron hole pair in the surface region, owing to the breaking of translational invariance. In strength of this damping effect depends susceptibility on shape of the electron density prifile[40]. For the Lang-Kohn profile and probably for real metals, the damping effects are so large that the MSP modes may easily be overlooked. Owing to the strong damping, direct observation of multipole surface Plasmon in the ATR experiment may be hard if not possible. On the other hand, it would be possible to excite their modes, in contract to the regular surface Plasmon, even at a perfectly flat surface by incident light become. They are related to peak in Im $\{d_\perp(\omega)\}$ i.e. in the absorptance. Therefore exication of MSP would lead to a reduced intensity of reflected light. Our result are shown in tables $5T_1$. There results are exact. For thick metallic layer on metal substrates, it is necessary to work with the exact (within HD) formula but for this surface layer and clean surfaces simplification are possible.

Table: Result of inverse reflectivity $1/|r|^2$ for k/k_x =0.005, 0.05 and 0.1 with $d_\perp(\omega)$ values for RPA in Chapter IV. Other results are from HD approximation with k_x/k_F =0.1 and $r_s=0.3\omega_p$ and $0.15\omega_p$

ω/ω_p	Result of $1/|r|^2$				
	k_x/k_F =0.005 $d_\perp(\omega)$RPA result	k_x/k_F =0.05 $d_\perp(\omega)$ RPA result	k_x/k_F =0.1 $d_{\|\|}(\omega)$ RPA result	HD with k_x/k_F =0.1 $r_s=0.3\omega_p$	HD with k_x/k_F =0.1 $r_s=0.15\omega_p$
0.50	0.0685	0.0546	0.0479	0.0328	0.0437
0.55	0.0324	0.0269	0.0186	0.0096	0.0586
0.60	0.0059	0.0038	0.0017	0.0027	0.1073
0.65	0.0214	0.0139	0.0106	0.0158	0.2786
0.70	0.1053	0.0844	0.0655	0.0986	0.3849
0.75	0.2565	0.1752	0.1234	0.1627	0.4268
0.80	0.4598	0.3218	0.3008	0.1238	0.5137
0.85	0.5985	0.4297	0.4039	0.1048	0.4032
0.90	0.6713	0.6227	0.5882	101478	0.3546
0.95	0.7532	0.7016	0.6379	0.2639	0.3045
1.00	0.8059	0.7586	0.7016	0.3486	0.4176

References

[1] F. Abeles, T. Lopez – Rios : Surf. Sci., 96 32 (1980
[2] A.R. Melnyk, K.M. J. Hassison : Phys. Rev. Lett. 21, 85 (1968) : Phys. Rev. B2, 835 (1970)
[3] 1 Lindau, P.O. Nilsson : Phys. Rev. Lett A 31, 352 (1970) : Physical scripta 3, 87 (1971)
[4] Kokempa, R.R. Gerhatts : Solid State Commun 53, 579 (1985)
[5] F. Fosstmann : Z. Physik 203, 495 (1967)
[6] P. Ascarelli, M. Cini : Solid State Commun : 18, 385 (1926)
[7] A. Equiluz, J.J. Quinn : Nuovo cimento, 39B, 828 (1977)
[8] A. Equiluz, S.C. Ying, J.J. Quinn : Phys, Rev B11, 2118 (1975)
[9] A. Equiluz, J.J. Quinn : Phys. Lett. A53, 151 (1975)
[10] A. Equiluz, J.J. Quinn : Phys. Rev, B14, 1347 (1976)
[11] A.J. Bennet : Phys. Rev B1.203 (1970)
[12] A. Euiluz, S.C. Yung, J.J. Quinn, Phys. Rev. B11, 2118 (1975)
[13] A. Euiluz, J.J. Quinn, Phys. Lett A53, 151 (1975)
[14] A. Euiluz, J.J. Quinn, Phys. Rev B14, 1347 (976)
[15] K.Kempa, R.R. Gerhardts : Solid Srare Commun 53, 579 (1985)
[16] C. Schwatz, W.L. Schaich : Phys. Rev B30, 1059 (1984)
[17] A. Bagchi, R.G. Barrera, A.K. Rajagopal:Phys. Rev B20, 4824 (1979)
[18] J.D.E. McIntyre, D.E. Asphes : Surf. Sci, 24, 417 (1971)
[19] J.E. Slipe : Phys. Rev. B22, 1589 (1980)
[20] A.J. Bennett : Phys. Rev. B1.203 (1970)
[21] L. Genzel, T.P. Martin, U. Kreibig : Z. Phys. B21, 339 (1975)
[22] J.C. Maxwell-Garnett : Phil. Trans. Roy. Soc. 203, 385 (1904) : 205, 237 (1906)
[23] A. Leibsely, B.N.J. Persson : J. Phys C : Solid State Phys 16, 5375 (1983)
[24] R. Rupin : Phys. Rev B11, 2871 (1975)
[25] A.D. Boardmann, B.V. Prajape Y.O. Nakanura : Phys. State. Sol (b) 75, 347 (1976)
[26] J.E. Inglesfield, EL. Wikborg : J. Phys F5, 1706 (1975)