Plasmonic pressure in profile-modulated and rough surfaces

N Noginova¹,², M LePain², V Rono¹, S Mashhadi¹, R Hussain³ and M Durach¹

¹ Norfolk State University, Norfolk VA, USA
² Georgia Southern University, Statesboro GA, USA
³ Intel Corporation, Hillsboro OR, USA

E-mail: nnoginova@nsu.edu

Keywords: plasmon drag effect, surface plasmon polaritons, plasmon-electron interaction

Abstract

The electromagnetic momentum loss approach is applied to predict the plasmon drag effect in profile-modulated gold films at different amplitudes of surface modulations. Direct proportionality of energy and momentum transfer in interactions of plasmons and free electrons in metal is proven to be valid for surfaces with relatively low height modulation amplitudes. An equivalent circuit model is discussed, which can provide qualitative description of the plasmon-induced electrical effects in modulated surfaces and surfaces with random roughness.

1. Introduction

Plasmon drag effect (PLDE) is a giant enhancement of photoinduced electric currents in metal films and nanostructures [1–9], associated with excitation of surface plasmon polaritons (SPPs). PLDE presents interest for various applications as it provides an opportunity for direct electronic monitoring of plasmonic elements in electronic and optoelectronic devices and sensors. From a fundamental point of view, the effect can be considered in terms of plasmon-assisted momentum transfer in light–matter interaction in metal [2]. In the first PLDE experiments [1, 3], strong enhancement of photoinduced currents were observed in flat gold and silver films under conditions of surface plasmon polariton resonance excited in Kretschmann geometry [10]. Significant photo-induced electric currents were reported in rough and nanostructured surfaces under direct illumination as well, with polarity and magnitude of electric signals dependent on angle of incidence, wavelength of illumination, light polarization, and nanoscale surface geometry [6–9].

Theoretical description of PLDE is based on the electromagnetic momentum loss approach. In a metal nanostructure, the rate of momentum transfer from field to electrons can be found as [2]

\[ \tilde{j}_L = \frac{i}{2} \text{Re} \left[ P_\alpha \partial_\alpha E^*_\alpha \right], \]

where \( P = \chi E \) is the polarization vector, \( E \) is the electric component of the optical field, \( \chi \) is the susceptibility of the material, \( i, \alpha = x, y, z \) and summation over \( \alpha \) is implied. It was recently shown [11] that this approach modified to take into account the electron thermalization time [12, 13], \( \tau_{\text{therm}} \) adequately describes strong electric currents induced under SPP resonance conditions in flat films. The theory was extended to surfaces with modulated profiles and multi-mode plasmonic excitations [11]. Under certain assumptions (a relatively small amplitude of surface modulation height, and laminar electric current), the electromotive force (emf) per unit length induced in a plasmonic structure can be presented as a sum of contributions from each plasmonic element, with the contribution proportional to the absorbed power \( Q_m \) and \( k \)-vector of each mode, \( k_m \), as

\[ U = \sum_m U_m = \frac{\tau_{\text{therm}}}{n_e} \frac{1}{\hbar} \sum_m \frac{\hbar k_m}{\omega} Q_m, \]

where \( \tau \) is the Drude collision time, and \( n_e \) is the electron density.

According to [11], strict numerical calculations (equation (1)) correctly predict the angular position, magnitude and polarity of the photoinduced currents experimentally observed in flat silver films [3] and a gold film with a sine-wave profile [11] at the SPP resonance conditions. However, in both cases, the exact shape of the
peak as well as additional off-resonance signals are not properly described by the theory; and this is tentatively attributed to the effect of roughness in experimental samples. Below, we provide details of our calculations for the sine-wave films, explore limitations of our theoretical approach [11] and validity of a simplified consideration (equation (2)). In addition we present a simple approach based on an equivalent circuit model, which provides qualitative description of the photoinduced emf in profile modulated and rough surfaces.

2. Theory. Strict calculations

The structure under consideration is a thin gold film of 60 nm thickness with a sine-wave profile, a periodicity \(d = 538 \text{ nm}\) and a modulation depth \(2h\), which was varied in theoretical simulations, see figure 1(a). The electromagnetic fields in a sine-wave structure are calculated based on the Chandezon method \([13]\) at the incidence angle, \(\theta\)

\[
k_{\text{pp}} = nG + k_x,
\]

where \(k_x = k \sin \theta\) is the projection of the optical \(k\)-vector on the \(x\) axis, \(G\) is the grating vector with a magnitude of \(G = 2\pi/d\), \(n\) is an integer. The SPP \(k\) vector closely corresponds to the estimations for flat films \([15]\)

\[
k_{\text{pp}} = k = \frac{1}{k_x} = \frac{\varepsilon_m / \varepsilon_d}{\sqrt{\varepsilon_m + \varepsilon_d}},
\]

where \(\varepsilon_m\) and \(\varepsilon_d\) are correspondingly dielectric permittivities for metal and air.

Reflectivity of the film, \(R(h, \theta, \omega)\) was calculated for the whole optical range for various amplitudes of the surface modulation, \(h = 0 – 50 \text{ nm}\), as a function of incidence angle, \(\theta\), and light frequency (see an example in figure 1(b)). Two SP branches are seen, which can be fitted with equation (3) with \(n = -1\) or \(+1\), corresponding to the plasmon propagating backward (against \(k_x\), low frequency branch), and forward (in the direction of \(k_x\), high frequency branch). These modes join each other at zero incidence angle and a frequency of \(\sim 2.15 \text{ eV}\), forming a standing plasmon wave with \(k_{\text{pp}} = G / \xi\).

In figure 1(c), the reflectivity \(R\), normalized to the reflectivity of the flat film, \(R_0\), is plotted as a function of frequency for different modulation amplitudes at \(\theta = 5^\circ\). Each curve demonstrates two SPP resonance dips and an additional small peak around \(2.15 \text{ eV}\) related to Rayleigh anomaly \([16]\). With an increase in the modulation amplitude, the resonance dips first become deeper and then broader with the further increase of \(h\). Figure 1(d) shows the spectral dependences of losses associated with the SPP excitation, \(P_{\text{SPP}}\), which were calculated as the difference between the reflected intensity from a flat gold film \((h = 0)\) and that at the conditions of the SPP resonance for a particular \(h\) for the same \(\theta\) and \(\omega\). At low \(h\) (10 and 20 nm), \(P_{\text{SPP}}\) decreases with the increase in the frequency demonstrating a small peak at \(\sim 2.15 \text{ eV}\). \(P_{\text{SPP}}\) is the highest at \(h \approx 30 \text{ nm}\) for most of the spectrum,

\[P_{\text{SPP}} = \binom{1}{1}(U_0^\text{r})(R_0^\text{r}) \text{ at SPR} = \text{mV/}\text{MW cm}^{-2}.
\]
indicating the most efficient SPP excitation. With the further increase in \( h \) the spectral dependence \( P_{SPP}(\omega) \) tends to, instead, increase with the increase of \( \omega \).

The plasmonic pressure force [2] acting on an electron, is determined by fields in metal as

\[
f_a = \frac{1}{2} \text{Re} \left\{ \chi (E_r \partial_r E_r^* + E_i \partial_i E_i^* + E_x \partial_x E_x^*) \right\}.
\]

(5)

The photoinduced emf is calculated by averaging the pressure force, \( f_a \), over the volume of the metal film as [11]

\[
\frac{U}{T} = \frac{\tau_{\text{therm}}}{\tau} \frac{1}{\kappa_{\text{a,b}}} \int_0^\ell f_a(z) \, dz,
\]

(6)

where \( I \) is the incident light intensity and \( L \) is the diameter of the illuminated spot.

The emf calculated in a flat film \((h = 0)\), \( U_0 \), is pure photonic drag [17], which is small and plays a role only at the blue part of the spectrum. The plasmonic emf, \( U - U_0 \), calculated at \( \tau_{\text{therm}} = 270 \, \text{fs} \), \( L = 2 \, \text{mm} \) and \( \theta = 5^\circ \) is plotted in figure 1(e). The polarities of \( U - U_0 \) at red and blue parts of the spectrum are opposite to each other, in each case corresponding to the drift of electrons in the direction of SPP propagation. The emf peaks change with the variation of \( h \) in similar way to the dips in the reflectivity (figure 1(c)).

For further analysis, the ratio of the peak emf at SPR conditions and maximum losses associated with SPR are plotted in figure 1(f) for the whole optical range. The curves are almost flat, corresponding to

\[C = \frac{1}{\ell} \frac{U - U_0}{\kappa_{\text{a,b}}} = 2.2 - 2.5 \, \text{mV} \, (\text{MW}^{-1} \, \text{cm}^{-2})\]

at small modulation amplitudes, increasing to \( \sim 3 - 3.5 \) with an increase in \( h \). They are negative at the red part of the spectrum, and positive at blue, corresponding to backward or forward plasmon drag. The polarity switches at \( \sim 2.15 \, \text{eV} \) where the standing SPP wave is excited, providing zero momentum to electrons. Near the switching point the curves with high \( h \) demonstrate a small peak.

The flatness of the curves indicates that the momentum transfer from plasmons to electrons is directly proportional to the energy transfer, which correlates well with the conclusions of the theory [11] predicting a direct proportionality between the plasmon induced emf and absorption at relatively small modulations of the metal surfaces \((h \ll d)\) and laminar electron flow. According to the results of the simulations, it holds up to amplitudes of \( 40 - 50 \, \text{nm} \) for our periodicity. In comparison with the experiment, the calculations correctly predict the magnitude, polarity and angular position of the PLDE peak observed experimentally in the gold film with \( h \sim 25 \, \text{nm} \) see, figure 3 in [11]). However, the presence of additional contributions (clearly seen at large incidence angles) needs to be taken into account.

3. Equivalent circuit model

While accounting for random roughness presents a challenge for exact numerical simulations, let us now make an attempt to discuss the photoinduced emf in the frame of the equivalent circuit model and test the possibility of explaining the signal observed at large angles with SPPs excited due to roughness. The goal of the consideration below is not to achieve full correlation with numerical calculations and experimental data, but rather to interpret the results and explore general tendencies. Consider a circuit consisting of a transmission line coupled through the loop with the resonance circuit consisting of two cavities having different resonance frequencies, see figure 2. The reflectivity from such a resonant circuit can be found as

\[|\Gamma|^2 = \frac{Z_2 - Z}{Z_2 + Z}^2,\]

(7)

where \( \Gamma \) is the complex reflection coefficient, \( Z_0 \) is the impedance of the transmission line, and \( Z \) is the equivalent input impedance [18]

\[
Z = i\omega L_0 + \frac{i\kappa_{a,b} \omega^2}{\omega^2_a - \omega^2 + \frac{i\kappa_{a,b} \omega^2}{Q_a}} + \frac{i\kappa_{b,b} \omega^2}{\omega^2_b - \omega^2 + \frac{i\kappa_{b,b} \omega^2}{Q_b}}.
\]

(8)

\[\kappa_{a,b} = \frac{\omega M_{a,b}}{L_{a,b}}.\]

(9)

Here \( M_{a,b}, L_{a,b} \) and \( Q_{a,b} \) are the mutual inductance and equivalent inductances of the line and the resonant circuits respectively, \( r_{a,b} \) and \( C_{a,b} \) are the equivalent resistance and capacitance of the resonant circuits, \( \omega_{a,b} \approx (L_{a,b} C_{a,b})^{-1/2} \), and \( Q_{a,b} = \omega_{a,b} L_{a,b}/r_{a,b} \). Energy exchange between incident light and the SPPs in optical experiments can be viewed in terms of the energy exchange between the transmission line and two resonant cavities corresponding to the two branches of SPR: the cavity ‘\( a \)’ describes the excitation of the forward SPP, and the cavity ‘\( b \)’ describes the ‘backward’ SPP. Coupling is provided with the sine-wave profile of the film. We assume \( \kappa_{a}/r_0 = \kappa_{b}/r_0 \sim h^{3/2}, \) which, in terms of an equivalent circuit, corresponds to the linear dependence of the mutual inductance on the modulation amplitude, \( M_{a,b} \sim h \). As shown below, this assumption well
corresponds to the results of the numerical calculations. Let us also take into account the fact that for the same incidence angle, the dip in reflectivity is broader for higher photon energies, figure 1(c), and assume that Q-factor of the cavities quickly decreases with the increase in the resonance frequency as $Q(E) = B/\omega_0^6$, where $B$ is the constant.

Equations (7)–(9) predict dips in the reflectivity at the resonance frequencies, figure 3(a). The curves are obtained assuming a predominately active resistance for the transmission line, $Z_0 = r_0 >> \omega L_0$, the resonance frequencies 2.015 and 2.285 eV, the Q-factors of 48 and 22.5, and various $\kappa/r_0$ proportional to $h^2$, where $h$ varies from 10 to 50 nm. As one can see, the equivalent circuit model predicts the results very similar to numerical simulations, including the initial increase in depth with the increase of $h$ from 10 to 30 nm, and broadening of the dip with a further increase of $h$. Variation of the incidence angle $\theta$ can be approximately described with the ‘tuning’ of the resonance frequencies of cavities. Varying both resonance frequencies toward each other with the point of coincidence at 2.15 eV, the spectral dependences of $1-|\Gamma|^2$ were calculated, figure 3(b). In similarity with...
As was observed in nominally flat films in the experiments with prisms [3]. Consider a random roughness with the Gaussian distribution

\[ F(G) = f_\delta \cdot \delta^{-1} \exp \left( -\frac{G^2}{\delta^2} \right), \]

where \( f_\delta \) is a constant, and \( \delta \) is the half-width of the distribution, figure 3(d). Substituting equation (13) to equation (12)

\[ U_r \propto \frac{\delta^2}{\pi^2} \exp \left[ -\frac{2\omega_0^2}{\delta^2} (\xi^2 + \sin^2 \theta) \right] \sinh \left[ \frac{4\omega_0^2 \xi \sin \theta}{\delta^2} \right], \]

while the power losses in such cavities are

\[ P_l \propto \frac{\delta^2}{\pi^2} \exp \left[ -\frac{2\omega_0^2}{\delta^2} (\xi^2 + \sin^2 \theta) \right] \cosh \left[ \frac{4\omega_0^2 \xi \sin \theta}{\delta^2} \right]. \]

In figure 3(e), the photoinduced emf associated with roughness is plotted for the various \( \delta \). At a relatively broad distribution, \( \delta > k_{\text{app}} \), the angular dependence is similar to the experimental dependences observed in the rough films [6], where the dependence \( U \propto \sin \theta \) is reported.

In figure 4 we make an attempt to fit our experiment [11] at 630/632 nm, making an assumption that the total change in reflectivity at high angles is due to additional SPPs excited in \( x \) or \(-x\) direction with the \( k \)-vector being the same as in flat films, \( \xi \sim 1 \). As one can see, the account for a roughness with the relatively narrow distribution, \( \delta/k_{\text{app}} \sim 0.5:1 \) can reasonably fit both the angular dependences of \( R \) and \( U \) in our sample. The possible source of such a distribution can be associated with the variation of the modulation height in the experimental sample. However, the quantitative fitting of the data for large incidence angles is still problematic even for such strong assumptions. One of the possible reasons of the discrepancy of the experiment and this particular fitting is the participation of plasmonic modes with high \( k \)-vectors excited at corrugated surfaces [19], which may have correspondingly higher contributions to the photoinduced voltage, \( U_1 \sim \xi \). Note that localized surface plasmons commonly associated with roughness are non-propagating and do not contribute to PLDE.
Another factor is the limitations of the simplified consideration for surfaces with relatively high slopes of the surface modulation. As seen from figures 1(f) and 3(c), the predictions of the exact numerical simulations and equivalent circuit model for high modulation amplitudes have different behavior around the switching point.

In conclusion, the PLDE in a thin gold film with a sine-wave profile was analyzed theoretically for various amplitudes of the surface modulations in a broad optical range. Both the numerical simulations based on modified electromagnetic momentum loss approach and the equivalent circuit model based on the simplified approach were shown to provide adequate description of the emf associated with propagating SPPs at small amplitudes of surface modulations. To take into account the effect of roughness, we considered contributions from additional plasmonic modes excited at rough surfaces as additional resonance cavities in the equivalent circuit and demonstrated that this can provide a qualitative picture of the effect. Limitations of the simplified consideration were discussed.

Acknowledgments

The work was partially supported by PREM grant no. DMR 1205457 and the Army Research Office (ARO) grant W911NF-14-1-0639. M D was supported by funds from the Office of the Vice President for Research & Economic Development at Georgia Southern University.

References

[1] Vengurlekar A and Ishihara T 2005 Appl. Phys. Lett. 87 091118
[2] Durach M, Rusina A and Stockman M I 2009 Phys. Rev. Lett. 103 186801
[3] Noginova N, Yakim A V, Soimo J, Gu L and Noginov M A 2011 Phys. Rev. B 84 035447
[4] Kurosawa H and Ishihara T 2012 Opt. Express 20 1561
[5] Kurosawa H, Ishihara T, Ikeda N, Tsuya D, Ochiai M and Sugimoto Y 2012 Opt. Lett. 37 2793
[6] Noginova N, Rono V, Bezares F J and Caldwell J D 2013 New J. Phys. 15 113061
[7] Akbari M, Onoda M and Ishihara T 2015 Opt. Express 23 823
[8] Ni X, Xiao Y, Yang S, Yu W and Zhang X CLEO: 2015, OSA 2015, # FW4E.1
[9] Noginova N, Rono V, Jackson A and Durach M CLEO: 2015, OSA 2015 # FTh3E.7
[10] Kretschmann E 1972 Opt. Commun. 6 185
[11] Durach M and Noginova N 2016 Phys. Rev. B 93 161406
[12] Farm W S, Storz R, Tom H W K and Bokor J 1992 Phys. Rev. Lett. 68 2834
[13] Sun C-K, Vallée F, Acioi L H, Ippen E P and Fujimoto J G 1994 Phys. Rev. B 50 15337
[14] Chandezon J, Raoult G and Maystre D 1980 J. Opt. 11 235
[15] Raether H 1988 Surface plasmons on smooth and rough surfaces and on gratings Springer Tracts in Modern Physics vol 111 (New York: Springer)
[16] Brevik I 1986 Phys. Rev. B 33 1058
[17] Maystre D 2012 Theory of Wood’s anomalies Plasmonics ed S Enoch and N Bonod (Berlin: Springer) pp 39–83
[18] Montgomery C G, Dicke R H and Purcell E M 1948 Principles of Microwave Circuits (New York: McGraw-Hill)
[19] Novotny L and Hecht B 2012 Principles of Nano-Optics 2nd edn (Cambridge: Cambridge University Press)