Surface Plasmon Amplification in Planar Metal Films

Dentcho A. Genov, Muralidhar Ambati, and Xiang Zhang

***Abstract*—The propagation and amplification of surface plasmon polaritons (SPPs) is studied at the interfaces between metals and active media. A permittivity renormalization technique is proposed and developed to obtain an explicit analytic expression for the critical gain required to achieve infinite SPP propagation length. A specific multiple quantum-well (MQW) system is identified as a prospective medium for demonstrating efficient SPP amplification at telecommunication frequencies. The proposed system may have a strong impact on a variety of photonic devices ranging from plasmonic nanocircuits, subwavelength transmission lines and plasmonic cavities to nanosized transducers.**

***Index Terms*—Multiple quantum wells (MQWs), planar metal films, surface plasmons.**

I. Introduction

THE CONCEPT of collective electron excitations, surface plasmon polaritons (SPPs), was first introduced by Ritchie fifty years ago [1]. Since then, a new scientific field, referred to as plasmonics, has rapidly developed resulting in numerous applications in bio-sensing [2], nanolithography [3], novel type of metal composites [4], superlenses and negative index materials [5]–[7]. Due to the prospective impact of surface plasmons on applied science, it is imperative that a straightforward analytic treatment should be made possible to describe both, the dispersion and propagation properties of the collective electronic states in optically passive and active media.

Stern was the first to derive the SPP dispersion at a single metal-dielectric interface [8]. The SPP's propagation properties were later studied theoretically and experimentally [9]. Extensive investigation of the SPPs in thin metal films, in the absence of intrinsic metal losses, was presented in [10], where approximate solutions were derived for the SPP dispersion. The attenuation of the surface waves was also studied for passive media on smooth [11] and rough [12] film surfaces. Recently, the interaction of surface plasmons with an active media has been theoretically discussed at a single metal-dielectric interface [13], [14] and in random composite materials [15], [16], with first experimental results showing SPP lasing [17] and effects of gain on SPP absorption [18]. Despite the large number of existing theoretical works, a reliable and explicit analytical solution of the SPP propagation in passive and active media that holds for broad frequency range and arbitrary film thicknesses has not been made available so far. In this paper, we introduce a permittivity renormalization technique that yields explicit solutions of the SPP's dispersion and propagation length for frequencies below resonance. Based on this new approach, we derive for the first time the explicit conditions for exciting SPPs with infinite propagation length, and an actual physical system is proposed to study this phenomenon. Possible applications in the emerging field of nanophotonics [19]–[21], and integration with the current CMOS technology are considered.

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II. Permittivity Renormalization Technique

Following a commonly applied procedure [9], we seek a solution of the Maxwell's equations in the form of a transversal (TM) surface wave propagating along a metal slab of thickness d and bounded in vertical z direction [see inset in Fig. 1(a)]. By restricting our analysis to a symmetric host environment and enforcing the boundary conditions at the metal interfaces, it is straightforward to obtain the SPP dispersion as in

$$k_{s,a}^2 = k_0^2 \frac{\varepsilon_m \varepsilon_d (\varepsilon_d - \rho_{s,a}\varepsilon_m)}{\varepsilon_d^2 - \rho_{s,a}\varepsilon_m^2} \tag{1}$$

where $k_0 = \omega/c$, $\rho_s = \tanh^2[(1/2)d\sqrt{k_s^2 - \varepsilon_m k_0^2}]$ and $\rho_a = \coth^2[(1/2)d\sqrt{k_a^2 - \varepsilon_m k_0^2}]$ correspond to two SPP modes with symmetric (s) and antisymmetric (a) tangential electric field profiles (with respect to $z = -d/2$ plane), and ε_m is the metal permittivity. The host permittivity $\varepsilon_d = n^2(1+i\Delta)$ is a complex function, with $\Delta > 0$ corresponding to an active and $\Delta < 0$ to a dissipative medium.

The transcendental equation (1) does not allow for an explicit solution of the SPP dispersion in the form $k = k(\omega)$ or $\omega = \omega(k)$. To overcome this problem, we propose an iterative method followed by the concept of quasi-metal. We begin by assuming an initial solution in the form of a plane wave, $k_{s,a}^{(0)} = nk_0$ and substituting $\rho_{s,a}^{(0)} = \rho_{s,a}(nk_0)$ in (1), we obtain an explicit first-order estimate $k_{s,a}^{(1)} = k_{s,a}^{(1)}(\omega)$ of the SPP dispersion. This procedure can be repeated to obtain higher order estimates $k_{s,a}^{(m)} = k_{s,a}^{(m)}[\omega, k_{s,a}^{(m-1)}(\omega)]$, but requires considerable mathematical effort. Fortunately, a highly accurate analytic representation can be derived based only on the first-order result. For that we introduce a quasi-metal permittivity $\varepsilon_m \to \bar{\varepsilon}_m(\omega, d)$ such that $k_{s,a}^{(1)} = k_{s,a}^{(1)}(\omega)$ reduces to the case of SPP at a single interface $k^2 = k_0^2\varepsilon_d\bar{\varepsilon}_m/(\bar{\varepsilon}_m + \varepsilon_d)$, which is easily solved explicitly [9], [10]. To carry out the proposed permittivity renormalization, it is essential to establish suitable representation of the metal permittivity. In the visible and infrared spectral ranges the bulk metal permittivity is well described with the Drude model $\varepsilon_m = n_b^2 - \omega_p^2/(\omega^2 - i\omega\omega_\tau)$, where n_b is a contribution due to bound electrons, ω_p is the plasma frequency and ω_τ is the relaxation rate. For noble metals, and $\omega < \omega_p$, we write $\varepsilon_m = \varepsilon_m' + i\varepsilon_m'' = (-1 + i\kappa)/\varepsilon$, where $\varepsilon = 1/|\varepsilon_m'| \ll 1$

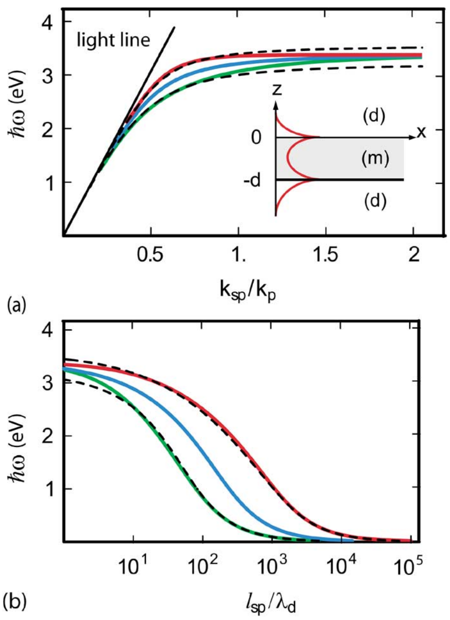

Fig. 1. $\omega - k_{\rm sp}$ (a) and $\omega - l_{\rm sp}$ (b) diagrams for silver-glass system, are calculated for metal slab with thickness $d = 40$ nm. The exact results (solid lines) and the PR expansion (dashed lines) are depicted for both, symmetric (green) and antisymmetric (red) SPP modes. The limiting case of a single silver-glass interface is depicted with blue line, and a schematic of the metal slab with the transversal SPP field profile is included as an inset. In all calculations, we use silver with experimentally fitted parameters: $\omega_p = 9.1$ eV, $\omega_c = 0.02$ leV, $n_b = 2.24$ [22], and $\lambda_d = \lambda/n$ is the wavelength in the host media.

and $\kappa = \varepsilon''{}_m/|\varepsilon'_m| \ll 1$. Using this representation we expand $\rho^{(0)}_{s,a}$ in series over κ. Keeping only the first-order terms we arrive at $\rho^{(0)}_{s,a} = r_{s,a}(1 + i\kappa\varphi_{s,a})$, where the modal functions $\varphi_s = -\varphi_a = (1/2)d\Lambda\mathrm{csch}((1/2)d\Lambda)\mathrm{sech}((1/2)d\Lambda)$ and $r_s = 1/r_a = \tanh^2((1/2)d\Lambda)$, are in general frequency dependant, since $\Lambda = k_p\sqrt{1+n^2\varepsilon}$ ($k_p = \omega_p/c$ is the plasma wave vector).

Substitution of $\rho^{(0)}_{s,a}$ in (1) reduces it to the case of a single interface between an insulator and a quasi-metal, with SPP wave vector $k_{\rm SP} = {\rm Re}k$ and propagation length $l_{\rm SP} = 1/2{\rm Im}k$ given by

$$k_{\rm SP} = n_{\rm SP}k_0, l_{\rm SP} = k_{\rm SP}^{-1}\frac{1 - n^2\bar{\varepsilon}}{\Delta + n^2\bar{\kappa}\bar{\varepsilon}} \tag{2}$$

where $n_{\rm SP} = n/\sqrt{1 - n^2\bar{\varepsilon}} > n$ is the SPP index of refraction, and the mode dependant quasi-metal permittivity $\bar{\varepsilon}_m = (-1 + i\bar{\kappa})/\bar{\varepsilon}$ is a function of the incident frequency and slab thickness

$$\begin{aligned} \bar{\varepsilon}_{s,a}(\omega, d) &= \varepsilon\frac{1 + n^2\varepsilon}{r_{s,a} + n^2\varepsilon}, \theta_s = r_s, \quad \theta_a = 1 \\ \bar{\kappa}_{s,a}(\omega, d) &= \frac{\kappa\varepsilon}{\bar{\varepsilon}_{s,a}}\frac{1 + \varphi_{s,a} + 2n^2\theta_{s,a}\varepsilon}{r_{s,a} + 2n^2\varepsilon}. \end{aligned} \tag{3}$$

Note, that the proposed permittivity renormalization preserves the asymptotical behavior in the limit of thick films ($d\Lambda > 1$), where the bulk metal properties are to be recovered; $\bar{\varepsilon}_{s,a} \to \varepsilon$ and $\bar{\kappa}_{s,a} \to \kappa$ (since $\lim_{d\to\infty}\varphi_{s,a} = 0$ and $\lim_{d\to\infty}r_{s,a} = 1$).

The accuracy of the analytical results is demonstrated in Fig. 1(a)–(b), where we have also numerically solved the transcendental equation (1). Overall, the analytical and exact numerical results match very well for frequencies up to the quasi-resonance ($n^2\bar{\varepsilon} = 1$). A better correlation is observed in the case of the asymmetric mode where the theory follows closely the exact numerical values throughout the entire propagation band except for a narrow range where the SPP's group velocity $v_g = \partial\omega/\partial k$ is negative. For real metals, those modes are highly dissipative and not suitable for application in SPP based transmission lines and active plasmonics devices [19]–[21], which is the main goal of this work.

III. Multiple Quantum-Well System for Surface Plasmon Amplification

Relying on the analytical theory we are now capable to explicitly study the interaction of the SPPs with an active media. An inspection of (2) clearly shows that provided a gain media ($\Delta < 0$) completely surrounds the metal slab, a SPP amplification, characterized by infinite propagation length, could be expected. To study this critical phenomenon, we consider a strain compensated InGaAs–InP multiple quantum well (MQW) positioned on both sides of a silver metal slab. Under operation far below saturation, we can introduce a linear material gain α_m and a SPP modal gain $\alpha_{\rm SP} = -nk_0\Delta = \alpha_m\Gamma - \alpha_{\rm abs}(1 - \Gamma)$, where $\alpha_{\rm abs}$ corresponds to absorption in the InP barrier layer. The electromagnetic confinement factor is given as $\Gamma = \sum_i \int_{z_i}^{z_i+w} |S_x(\omega, z)|dz/\int_0^\infty |S_x(\omega, z)|dz$, where S_x is the time averaged x-component of the SPP power flux, z_i is the position of the ith QW in relation to the metal slab and w is its thickness. For a periodic arrangement of the MQWs with period b, it is straightforward to obtain $\Gamma_{s,a} = (1 - e^{2w\Lambda_d^{s,a}})/(1 - e^{2b\Lambda_d^{s,a}}) \approx w/b$, where $\Lambda_d^{s,a} = {\rm Re}\sqrt{k_{s,a}^2 - \varepsilon_d k_0^2}$ and we have taken into account that for frequencies below resonance $w\Lambda_d^{s,a} < b\Lambda_d^{s,a} \ll 1$, regardless of the investigated SPP mode. Clearly, dense packing of the MQW system favors higher modal gain. However, with decreasing spacing between the individual QW and their distance to the metal film, a considerable decrease in performance could be expected due to reduced carrier confinement and enhanced spontaneous emission [23]–[26]. Specifically, exciton relaxation into lossy surface waves in the metal will result in increase of threshold carrier injection rates necessary to maintain the same carrier concentration.

The material gain α_m is calculated by implementing the time-dependant perturbation theory, in which a Lorentzian carrier lifetime broadening $\ell_\gamma(\omega)$ is used

$$\begin{aligned} \alpha_m(\omega) = C_{\rm SP}\frac{2}{V} \\ \cdot\sum_{\vec{k}_v}\sum_{\vec{k}_c} M\delta_{\vec{k}_c,\vec{k}_v+\vec{k}_{\rm SP}}\ell_\gamma(E_c - E_v - \hbar\omega)(f_c - f_v) \end{aligned} \tag{4}$$

where $C_{\rm SP} = e^2\pi/c\omega\varepsilon_0 n_{\rm SP}m_0^2$, with c the speed of light in vacuum, ε_0 the permittivity of free space, m_0 the mass of an electron and e its charge, $M = |\hat{g}_{\rm SP}\cdot\vec{p}_{\rm vc}|^2$ the transition matrix element, $\hat{g}_{\rm SP} = \vec{A}_{\rm SP}/|\vec{A}_{\rm SP}|$ is a unit vector corresponding to the SPP vector potential $\vec{A}_{\rm SP} = (n\omega)^{-2}\vec{\nabla}\times\vec{H}_{\rm SP}$, $E_{c,v}$ and $\vec{k}_{c,v}$ are the conduction and valance electron energies and moments, respectively.

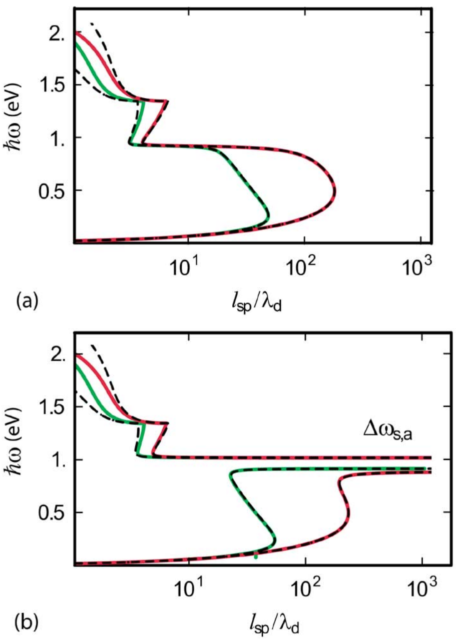

Fig. 2. $\omega - l_{\rm SP}$ diagrams for silver slab of thickness $d = 40$ nm, surrounded by MQW structure. The solid lines—anti-symmetric (red) and symmetric (green)—correspond to solution based on the exact relationship (1), while the permittivity renormalization is depicted with dashed black lines. For the unpumped case (a), strong damping is observed at low and high frequencies (compared to Fig. 2(b)). Injection of carriers, sheet carrier density $n_{2d} = 1.5 \times 10^{12}\text{cm}^{-2}$, (b) results in inversion of the MQW and formation of a frequency bands, $\Delta\omega_a = 0.13$ eV and $\Delta\omega_s = 0.1$ eV, where the SPPs are strongly amplified. The SPP modal parameters at peak gain are $n^S_{\rm SP} = 1.107 n_r, n^S_{\rm SP} = 1.034 n_r, \Gamma_s = 0.396$, and $\Gamma_a = 0.4052$.

The Fermi–Dirac population inversion factor $f_c - f_v > 0$, introduces an additional line broadening due to a finite temperature and band occupation. It is important to note that (4) differs from the gain in conventional laser systems in two important ways. First, the material gain α_m is reduced by a factor $n/n_{\rm SP}$ as compared to unbound plane wave propagation in the MQW. Second, a decrease in peak gain is expected due to the SPP polarization, which at low frequencies is predominantly TE in respect to the QW, but has a substantial longitudinal (TM) component at resonance.

For InGaAs–InP MQW with small well sizes ($w > 10$ nm) and at low temperatures $(\mathrm{T} = 0\mathrm{K})$ only the $e1 - hh1$ transition contributes to gain. The calculated SPP propagation length with and without gain is presented in Fig. 2. In these calculations, we set the characteristic parameters of the MQW as $w = 5$ nm and $b = 12$ nm, which defines the transition frequency as $\hbar\omega_{e1-hh1} = 0.92$ eV, and the exciton damping is taken from experimental data $\gamma \approx 5$ meV [27], [28]. For the unpumped system (Fig. 2a), and for frequencies above ω_{e1-hh1} and the InP bandgap ($\hbar\omega_g^{\rm InP} = 1.34$ eV), the SPPs are strongly attenuated by absorption in the QW and the barrier layer. Additionally, in the low frequency range, a strong decay for the long range surface plasmons is observed due to alloy scattering events and carrier-carrier interactions. With increasing temperature those two scattering processes will be supplemented by carrier-phonon interactions which may lead to further substantial increase in the SPP attenuation [27], [28].

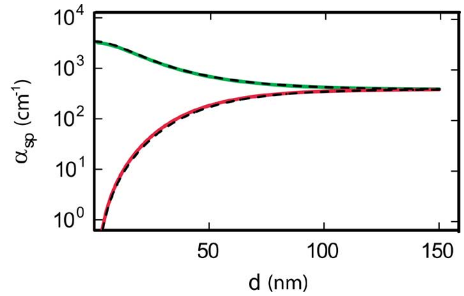

Fig. 3. Critical modal gain versus film thickness d d, at $\lambda = 1.3\ \mu$m ($h\omega = 0.92$eV) The numerical estimates based on (1) (solid lines) are compared to the explicit result (5) (dashed lines). The critical gain increases with decreasing film thickness for the symmetric SPP (green) and sharply decreases in the case of the antisymmetric (red) SPP mode.

Clearly, utilizing semiconductor materials, including MQWs, in conjunction with metallic systems for SPP related application is of no practical use as long as the system is not pumped. However, an entirely different picture arises when the MQW is inverted by injection of carriers. In general, the threshold sheet carrier density depends on the position of the QW in relation to the metal surfaces. Since most of the SPP energy flows in sheet of average thickness $t_{s,a} = 1/2\Lambda_d^{s,a}$, the population inversion is depleted predominantly by Auger recombination and spontaneous emission into SPPs free space modes. At low temperature the injection current density required to maintain sheet carrier density, $n_{2D} = 1.5 \times 10^{12}\text{cm}^{-2}$, is J >300 A/cm^2 [26], [28]. At this injection rates, the peak modal gains $\alpha^a_{SP,\max} = 5020\ \text{cm}^{-1}$ and $\alpha^s_{\rm SP,max} = 4586\ \text{cm}^{-1}$ are established. Such levels of amplification are sufficient for a frequency bands $\Delta\omega_{s,a}$ to occur [see Fig. 2(b)] where the SPP's attenuation is entirely compensated. Note that the MQW structure contributes to SPP amplification in only a narrow frequency band around the transition frequency of QW. The frequency amplification bands strongly depend on the intrinsic metal loss, investigated SPP mode and carrier injection rate. Since the energy of the asymmetric SPP is distributed mostly throughout the MQW and less inside the metal, the damping is lower and its frequency amplification band is broader, $\Delta\omega_a > \Delta\omega_s$, than that of the symmetric SPP.

IV. Critical Modal Gain to Compensate for Intrinsic Losses in Silver Metal

As discussed in the previous section, a crucial factor that determines the SPP amplification is the peak modal gain, which is fixed at low temperatures, but substantially decreases at ambient conditions. To study the critical modal gain $\alpha^{s,a}_{\rm SP} = -nk_0\Delta^{s,a}_c$ at which an infinite SPP propagation could be achieved, we numerically solve (1) by setting $\mathrm{Im}k^{s,a}_{\rm SP}(\omega_{e1-hh1}, \Delta^{s,a}_c, d) = 0$. The results are presented in Fig. 3, where a wide range of metal thicknesses have been studied. In the case of thick films, the SPP modes decouple and a modal gain $\alpha^0_{\rm SP} = -nk_0\Delta_0 = 430\ \text{cm}^{-1}$ is sufficient to provide amplification. With the decrease of the slab thickness, a split in dispersion relation is observed with the symmetric SPP mode necessitating much higher modal gain.

The explicit condition to achieve surface plasmon amplification is easily derived from (2). Specifically, a complete compensation of loss is expected for $\Delta = \Delta_c^{s,a} = -n^2\bar{\varepsilon}_{s,a}\bar{\kappa}_{s,a}$, which at low frequencies ($\varepsilon \ll 1$) simplifies as

$$\Delta_c^{s,a} = \Delta_0 \frac{1 + \varphi_{s,a} + 2n^2\theta_{s,a}\omega^2/\omega_p^2}{r_{s,a} + 2n^2\omega^2/\omega_p^2} \tag{5}$$

where $\Delta_0 = -n^2\varepsilon\kappa \approx -n^2\omega\omega_\tau/\omega_p^2$ is the critical value for a single metal-dielectric interface [13]. The approximate result (5) predicts virtually identical peak gains as those obtained from the direct numerical solution (see Fig. 3). It also provides important new insights into the problem. For instance, in the case of thin metal slabs, it predicts an asymptotic behavior of the critical gain, which for the asymmetric SPP mode diminishes with the film thickness $\lim_{d\to 0} nk_0|\Delta_c^a| = 2k_0n^5\kappa\varepsilon^2\tanh^2((1/2)k_pd) \approx 1940 \times (k_0d)^2$ cm^{-1}, whereas for the symmetric SPP reaches a maximum value $\lim_{d\to 0} nk_0|\Delta_c^s| = nk_0\kappa \approx 3200^{-1}$. As critical modal gain depends linearly on the metal losses, additional losses due to surface roughness of thin metal films can be accounted through modification of the metal permittivity $\kappa^{s,a}$ [9]. Although the required modal amplification, for the entire range of metal thicknesses, may be achieved in an actual device, it is clear that transmission lines based on SPP are most feasible for thin metal films and operation under asymmetric mode excitation. For instance, a silver slab of thickness $d = 25$ nm requires a moderate modal gain of $\alpha_{\mathrm{SP}}^a = 51$ cm^{-1} and threshold current density of $J_{\mathrm{th}} > 1$ A/cm^2 to achieve infinite SPP propagation, a value well below what has been demonstrated in practice [28].

V. Conclusion

To conclude, by introducing the concept of a quasi-metal, we have obtained an explicit solution of the SPP dispersion in metal films of arbitrary thickness. A critical gain for achieving infinite SPP propagation has been derived, and a strain compensated InGaAs–InP MQW system is proposed as an active media for direct demonstration of SPP amplification at telecommunication frequencies. The proposed theory can be easily extended to multilayer system consisting of metal and dielectric layers, which could be used to develop and optimize novel SPP based subwavelength sized transmission lines, interconnects and nanodevices compatible with the current in-plane CMOS technology.

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