Extraction efficiency of highly confined surface plasmon-polaritons to far-field radiation: an upper limit

Jaewoong Yoon[1], Seok Ho Song[1*], and Jin-Ha Kim[2]

[1]*BK21 Center in Department of Physics, Hanyang University, Seoul 133-791, Republic of Korea*
[2]*Corporate R & D Center, Samsung Electro-Mechanics Co, Ltd., Suwon 443-803, Republic of Korea*
**Corresponding author: shsong@hanyang.ac.kr*

http://optics.hanyang.ac.kr

Abstract: We propose a unique method determining an upper limit of extraction efficiency of the surface plasmon-polaritons (SPPs), E_{SP}, which are highly confined on a corrugated metal surface. The method is based on measurement of the spectral bandwidth of a grating-induced absorption spectrum as a function of metal dielectric constant. After finding the fact that E_{SP} exhibits an extremely linear relationship with the collision frequency Γ of metal over a SPP band below the surface plasmon frequency, an upper limit of E_{SP} can be determined by an asymptotic estimation as $\Gamma \rightarrow 0$ for total decay rates of the confined SPPs. Our method based on the bandwidth measurement is inherently free from the ambiguity and underestimation difficulties pertaining to the previous prism-coupling approaches for E_{SP} estimation. It will also be quite applicable for evaluating SPP-mediated light-emitting diodes (LEDs) of which total external efficiency is dominantly restricted by the upper limit of E_{SP}. Especially for the case when SPP excitation probability approaches unity, the proposed method would excellently figure out the maximum realizable external efficiency of SPP-mediated LEDs.

OCIS codes: (240.6680) Surface plasmons; (230.3670) Light-emitting diodes.

References and links

1. W. L. Barnes, A. Dereux, and T. W. Ebbesen, "Surface plasmon subwavelength optics," Nature **424**, 824-830 (2003).
2. E. Ozbay, "Merging photonics and electronics at nanoscale dimensions," Science **311**, 189-193 (2006).
3. R. A. Amos and W. L. Barnes, "Modification of the spontaneous emission rate of Eu^{3+} ions close to a thin metal mirror," Phys. Rev. B **55**, 7249-7254 (1997).
4. W. L. Barnes and P. T. Worthing, "Spontaneous emission and metal-clad microcavities," Opt. Commun. **162**, 16-20 (1999).
5. I. Gontijo, M. Boroditsky, E. Yablonovitch, S. Keller, U. K. Mishra, and S. P. DenBaars, "Coupling of InGaN quantum-well photoluminescence to silver surface plasmons," Phys. Rev. B **60**, 11564-11567 (1999).
6. J. Vuckovic, M. Loncar, and A. Scherer, "Surface plasmon enhanced light-emitting diode," IEEE J. Quantum Electron. **36**, 11331-1144 (2000).
7. W. L. Barnes, "Electromagnetic crystals for surface plasmon polaritons and the extraction of light from emissive devices," J. Lightwave Technol. **17**, 2170-2182 (1999).
8. P. A. Hobson, A. A. E. Wasey, I. Sage, and W. L. Barnes, "Role of surface plasmons in organic light-emitting diodes," IEEE J. Sel. Top. Quantum Electron. **8**, 378-386 (2002).
9. A. Neogi, C.-W. Lee, H. O. Everitt, T. Kuroda, A. Tackeuchi, and E. Yablonovitch, "Enhancement of spontaneous recombination rate in a quantum well by resonant surface plasmon coupling," Phys. Rev. B **66**, 153305 (2002).

#89163 - $15.00 USD Received 29 Oct 2007; revised 27 Dec 2007; accepted 8 Jan 2008; published 16 Jan 2008

10. K. Okamoto, I. Niki, A. Shavartser, Y. Narukawa, T. Mukai, and A. Scherer, “Surface-plasmon-enhanced light emitters based on InGaN quantum wells,” Nat. Mat. **3**, 601-605 (2004).
11. K. Okamoto, I. Niki, A. Scherer, Y. Narukawa, T. Mukai, and Y. Kawakami, "Surface plasmon enhanced spontaneous emission rate of InGaN/GaN quantum wells probed by time-resolved photoluminescence spectroscopy," Appl. Phys. Lett. **87**, 071102 (2005).
12. G. Sun, J. B. Khurgin, and R. A. Soref, "Practicable enhancement of spontaneous emission using surface plasmons," Appl. Phys. Lett. **90**, 111107 (2007).
13. J. Yoon, K. Choi, D. Shin, S. H. Song, H. S. Won, J. H. Kim, and J. M. Lee, "Enhanced external efficiency of inGaN/GaN quantum well light-emitting diodes by mediating surface plasmon-polaritons," J. Korean Phys. Soc. **50**, 1009-1017 (2007).
14. J. Moreland, A. Adams, and P. K. Hansma, "Efficiency of light emission from surface plasmons," Phys. Rev. B **25**, 2297-2300 (1982).
15. P. T. Worthing and W. L. Barnes, "Efficient coupling of surface plasmon polaritons to radiation using a bi-grating," Appl. Phys. Lett. **79**, 3035-3037 (2001).
16. P. T. Worthing and W. L. Barnes, "Coupling efficiency of surface plasmon polaritons to radiation using a corrugated surface; angular dependence," J. Mod. Opt. **49**, 1453-1462 (2002).
17. S. Park, G. Lee, S. H. Song, C. H. Oh, and P. S. Kim, "Resonant coupling of surface plasmons to radiation modes by use of dielectric gratings," Opt. Lett. **28**, 1870-1872 (2003).
18. A. Giannattasio, I. R. Hooper, and W. L. Barnes, "Dependence on surface profile in grating-assisted coupling of light to surface plasmon-polaritons," Opt. Commun. **261**, 291-295 (2006).
19. E. Kretschmann and H. Raether, "Radiative decay of non-radiative surface plasmons excited by light," Z. Naturforsch. **23A**, 2135-2136 (1968).
20. A. Otto, "Excitation of nonradiative surface plasma waves in silver by the method of frustrated total reflection," Z. Phys. **216**, 398-410 (1968).
21. H. Raether, *Surface plasmons on smooth and rough surfaces and on gratings* (Springer-Verlag, Berlin, 1988).
22. R. W. Wood, "On a remarkable case of uneven distribution of light in a diffraction grating spectrum," Phil. Mag. **4**, 396 (1902).
23. U. Fano, “The theory of anomalous diffraction gratings and of quasi-stationary waves on metallic surfaces (Sommerfeld’s waves),” J. Opt. Soc. Am. **31**, 213-222 (1941).
24. D. Maystre, M. Neviere, and P. Vincent, “On the general theory of anomalies and energy absorption by diffraction grating and their relation with surface waves,” Optica Acta **25**, 905-915 (1978).
25. A. Sharon, G. Glasberg, D. Rosenblatt, and A. A. Friesem, “Metal-based resonant grating waveguide structures,” J. Opt. Soc. Am. A **14**, 588-595 (1997).
26. M. G. Moharam and T. K. Gaylord, “Diffraction analysis of dielectric surface-relief gratings,” J. Opt. Soc. Am. **72**, 1385-1392 (1982).
27. L. D. Landau, E. M. Lifshitz, and L. P. Pitaevskii, *Electrodynamics of continuous media 2nd Ed.*, Translated by J. B. Sykes, J. S. Bell, and M. J. Kearsley, (Elsevier Butterworth-Heinmann, Oxford, 1984).
28. E. D. Palik, *Handbook of Optical Constants of Solids II* (Academic Press, San Diego, 1998).
29. J. C. Maxwell-Garnett, “Colours in metal glasses and in metallic films,” Philos. Trans. R. Soc. London **203**, 385-420 (1904).
30. J. A. McKay and J. A. Rayne, "Temperature dependence of the infrared absorptivity of the noble metals," Phys. Rev. B **13**, 673-685 (1976).
31. M.D. Tillin and J. R. Sambles, “Phonon assisted absorption in thin Ag films using surface plasmon-polaritons,” J. Phys.: Condens. Matter **2**, 7055-7059 (1990).

1. Introduction

Surface plasmon-polaritons (SPPs) have been accepted as an important subject in optical physics and technology because of their ability to interact strongly with nanoscopic objects [1,2]. In particular, use of SPPs for emissive devices such as blue or green light-emitting diodes (LEDs) is recently arousing special interests. Since theoretical and experimental results confirming that the internal quantum efficiency of an emitter can be remarkably enhanced by mediating SPPs in the spontaneous emission (SE) process [3-6], it is now expected that SPPs may provide an efficient way to a *better* LED, which is brighter, faster, and more directional [7-11]. For a SPP-mediated LED, light-emission is a two-stage process. On the first stage, SPPs efficiently gather energy from excited radiators with the help of large photonic mode density and tightly confined electromagnetic fields [4,9]. It is well known that this process is

#89163 - $15.00 USD Received 29 Oct 2007; revised 27 Dec 2007; accepted 8 Jan 2008; published 16 Jan 2008

so efficient that SE into SPPs largely dominates other energy transfer processes, such as SE into photons or phonons for appropriate device geometries [9-11]. The second stage is an extraction of light from the emitted SPPs by scattering them on a corrugated or structured surface. But the efficiency for the second stage has not been clearly verified so far. It is crucial for a *better* SPP-mediated LED to efficiently extract the excited SPPs to far-field radiation. It is evident that extraction efficiency of the excited SPPs, E_{SP}, should limit the total external efficiency of a LED especially for the case when the SE rate is highly enhanced under the surface-plasmon resonance (SPR) condition [12,13]. If the SPP excitation probability approaches unity, enhancement of the SE rate becomes enormous and the total external efficiency converges solely on E_{SP} [13]. Therefore, it is worth addressing the fundamental limit of the E_{SP} when designing a SPP-mediated LED.

Many experimental and theoretical works were conducted in order to determine E_{SP} precisely [14-18]. Most of these works were based on attenuated total-internal reflection (ATR) method to excite SPPs, and the amount of E_{SP} obtained was in the range of 50 ~ 80%. The ATR-coupling configurations like the Kretchmann and Otto types are quite useful for SPP excitation [19,20], but inherently possess some difficulties in exact determination of E_{SP} when a certain scattering structure like a diffraction grating is used for SPP extraction: There is an ambiguity in estimating the portion of incident power absorbed by the SPPs. Even at the reflectance minimum value of R_0 at a surface-plasmon resonance (SPR) angle, it is unclear whether the absorbed power is 1-R_0 or R_0'-R_0 where R_0' is a maximum reflectance estimated from the SPR curve. In the papers of Worthing et al [15,16], this ambiguity was not clarified and E_{SP} was given by 'some value' between $T_{-1}/(1-R_0)$ and $T_{-1}/(R_0'-R_0)$, where T_{-1} is the transmittance into the first-order diffraction. Another difficulty comes from leakage radiation of the SPPs back into the prism. It was already pointed out by Raether that an effect of surface roughness for SPP coupling can be reduced down to about half of its maximum efficiency, because the decay rate into leakage radiation is nearly the same as that of the ohmic decay when the SPR absorption is appreciable [21]. Therefore, there should be an underestimation of the role of gratings in E_{SP} measurement by ATR coupling. Moreover, the magnitude of SPP wavevectors in ATR coupling has to be practically limited by $k_{||} \le n_{prism} k_0$, where k_0 is a vacuum wavenumber. The refractive index of a commercialized prism is normally smaller than 2.0 over a visible frequency range, thus the ATR methods may not be adequate to excite such a highly confined SPP that provides not only huge enhancement in SE rate of a radiator but also nanoscale confinement in plasmonic integrated optics.

In this letter, we propose a unique method to determine an upper limit of E_{SP}. Our method is based on measuring the spectral bandwidth of a grating-induced SPR absorption spectrum as a function of ε_i of a metal dielectric constant, $\varepsilon_m=\varepsilon_r+i\varepsilon_i$. It is well known from the Wood's anomaly that the resonant excitation of a SPP reveals a Fano-type resonant diffraction in a Lorentzian absorption line [22-25] and that the half-width at half maximum (HWHM) of the absorption spectrum can be regarded as the total (internal + external) decay rate, $\gamma_t(\varepsilon_i)$, of an excited SPP. We have found from a calculation based on the rigorous coupled-wave analysis (RCWA) that [26], when $|\varepsilon_r| \gg |\varepsilon_i|$, $\gamma_t(\varepsilon_i)$ has an extremely linear relationship with the collision frequency Γ of the metal. Note that $|\varepsilon_r| \gg |\varepsilon_i|$ is valid over a SPP band below the surface plasmon frequency of most noble metals. Then we have finally found that an upper limit of E_{SP} can be determined by the asymptotic estimation of $lim_{\gamma \to 0}\, \gamma_t(\varepsilon_i)$. The proposed method based on the bandwidth measurement is inherently free from the ambiguity and underestimation difficulties pertaining to previous methods, such as the ATR-coupling. It can also be applicable to highly confined SPPs excited in a SPP-mediated LED in order to figure out the maximum realizable external efficiency.

#89163 - $15.00 USD Received 29 Oct 2007; revised 27 Dec 2007; accepted 8 Jan 2008; published 16 Jan 2008

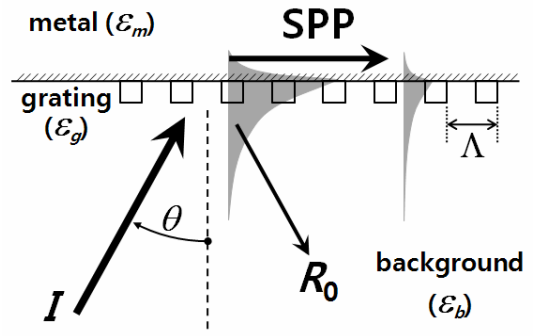

Fig. 1. Schematic of a system under consideration.

2. Method of E_{SP} estimation and its applicability to highly confined SPPs

Here we describe the detailed procedure and the practical validity of our bandwidth-measurement method for finding an upper limit of E_{SP}. After further numerical analysis demonstrating its successful application to a highly confined SPP, a consistency between the ATR and the proposed bandwidth-measurement methods is confirmed along with a definite evidence of the underestimation of E_{SP} in all previous results based on ATR coupling in sec. 3. A situation is assumed that light is made incident on a metal surface with a dielectric diffraction grating as shown in Fig. 1. The metal and the background dielectric are semi-infinite, and the diffraction grating with period Λ is in between them. The dielectric constant of each part is designated in the nearby parenthesis. At a SPR condition that

$$\sqrt{\varepsilon_b}\, k_0 \sin\theta = \pm k_{SP}(\omega) - 2\pi q / \Lambda \quad (q\text{: integer}), \tag{1}$$

the SPP excited by resonant coupling with the q-th order diffracted wave dissipates its energy into two distinct decay channels: One is internal (ohmic) decay into heat in the metal and the other is radiation decay into diffracted waves propagating away from the interface to the background dielectric. The resonant excitation of SPPs by the gratings reveals a Fano-type diffraction behavior with a Lorentzian absorption spectrum [22-25]. Accordingly, the absorption spectrum of the SPP resonance structure in Fig. 1 can be written by a series of Lorentzian resonances as

$$A(\theta,\omega) = A_b + \sum_q \frac{C_q}{[\omega - \omega_q(\theta)]^2 + \gamma_q(\theta)^2}, \tag{2}$$

where $\omega_q(\theta)$ is a frequency satisfying Eq. (1) at a certain incidence angle θ and $\gamma_q(\theta)$ a half-width at half maximum (HWHM). The series of Lorentzian resonances are in general attributed to the excitation of each damping mode with a complex eigenfrequency of $\Omega = \omega - i\gamma$, where ω is the resonance frequency and γ is the decay rate. Thus it is reasonable to assign $\omega_q(\theta)$ and $\gamma_q(\theta)$ to the q-th order resonance frequency and the q-th order total decay rate, respectively, satisfying the SPR condition of Eq. (1). The total decay rate is expressed by sum of the internal and radiation decay rates as $\gamma_q(\theta) = \gamma_{int} + \gamma_{rad}$. If we can measure γ_{int} and γ_{rad} separately, the probability of the radiative decay will be obtained by γ_{rad} /($\gamma_{int} + \gamma_{rad}$) and this value should be an upper limit of E_{SP}.

#89163 - $15.00 USD Received 29 Oct 2007; revised 27 Dec 2007; accepted 8 Jan 2008; published 16 Jan 2008

Now how can we measure γ_{int} and γ_{rad} separately? Dependences of γ_{int} and γ_{rad} on ε_i are quite different to each other. For the case when $|\varepsilon_r| \gg |\varepsilon_i|$, which is a reasonable assumption over a SPP band below the surface plasmon frequency of most noble metals, we can expect that the SPP mode has a field distribution nearly independent on ε_i. This independency of SPP modal fields implies that γ_{int} is linearly proportional to ε_i as can easily be confirmed by Poynting's theorem in lossy dispersive media [27], while γ_{rad} determined mainly by the index contrast and the depth of the grating is kept constant on ε_i variation. Consequently, under the condition that $|\varepsilon_r| \gg |\varepsilon_i|$, the total decay rate or the HWHM of SPR absorption line can be written as

$$\gamma_q(\varepsilon_i) = \gamma_{rad} + \gamma_{int}^{(r)}[\varepsilon_i / \varepsilon_i^{(r)}], \tag{3}$$

where $\varepsilon_i = (\Gamma/\omega)(1-\varepsilon_r)$ under the Drude model for a metal with a collision frequency Γ, $\varepsilon_i^{(r)}$ is a reference value of ε_i at a certain reference temperature, and $\gamma_{int}^{(r)}$ is the ohmic decay rate at $\varepsilon_i = \varepsilon_i^{(r)}$. The collision frequency can be reduced by lowering temperature, because the relaxation time of conduction electrons, $\tau = 1/\Gamma$, is getting longer as thermally excited phonons calm down at a lower temperature. The γ_{rad} is therefore given by the residual value of $\gamma_q(\varepsilon_i)$ when ε_i approaches zero, and finally E_{SP} at ω_q can be determined by

$$E_{SP} = \frac{\lim_{\Gamma \to 0} \gamma_q[\varepsilon_i(\Gamma)]}{\gamma_q[\varepsilon_i^{(r)}]}. \tag{4}$$

We confirm this supposition by RCWA calculation of SPP absorption spectra. Assume that $\varepsilon_b = 1$, $\varepsilon_g = 2.25$ (0.5 in filling ratio and 100 *nm* in depth), and the metallic half-space is an Ag film (1 μm in thickness) with $\varepsilon_m^{(r)} = -15.8639 + 1.0774i$ at vacuum wavelength of 632.8 *nm* [28]. At room temperature, the value of $\varepsilon_m^{(r)}$ is fitted to a Drude model in which the plasma frequency $\omega_p = 1.2257 \times 10^{16}$ *rad./s* and the collision frequency $\Gamma^{(r)} = 1.9031 \times 10^{14}$ *rad./s*. Figure 2(a) shows the 0th order reflectance (R_0) for Λ = 632.8 *nm*. Please note that the frequency ω and in-plane wavevector $k_{||}$ are scaled by those corresponding to a light with vacuum wavelength of 1 μm, and the units ($2\pi c/\mu m$ and $2\pi c/\mu m$) will be omitted hereafter. Several sharp SPR-absorption lines for $q = \pm 1$, ± 2, and ± 3 satisfying Eq. (1) can easily be recognized. The crossing points between the absorption boundaries indicate Bragg conditions of the excited SPPs. The straight edges which are parallel to the light lines at $\theta = \pm 90°$ showing sudden changes in R_0 correspond to the critical angle at which any diffracted order becomes an evanescent wave. Figure 2(b) shows the absorption spectrum (solid line) at normal incidence of $\theta = 0°$. R_0 (dashed curve) and $R_{\pm 1}$ (dot-dashed) are presented for comparison. Note that the absorption curve, A, comprises two well-defined Lorentzian SPR peaks at $\omega = 1.36$ (λ_0 = 735.3 *nm*) and 2.34 (λ_0 = 427.4 *nm*) which correspond to Bragg conditions of $q = \pm 1$ and ± 2, respectively, and that the q-th order diffraction of the excited SPP makes radiation toward the surface normal direction.

Effect of reducing the value of ε_i on the total absorption is shown in Fig. 2(c). The peak value at $\omega = 1.36$ ($q = \pm 1$) decreases with Γ and they shift within 0.4% from its original SPR frequency at $\Gamma = \Gamma^{(r)}$. The total decay rates γ_1 depicted in Fig. 2(d) are determined by least-square fitting of the absorption spectra to Lorentzian functions in Eq. (2). The filled squares are obtained from the RCWA calculation and the open square at $\Gamma / \Gamma^{(r)} = 0$ is a linearly extrapolated value. The total decay rate goes down linearly along with the solid line given by $\gamma_1 = [6.08 + 5.64\, \Gamma / \Gamma^{(r)}] \times 10^{-3}$. As expected, the total decay rate estimated from its linearity on Γ is well fitted to the value obtained by the RCWA within a maximum discrepancy of

#89163 - $15.00 USD Received 29 Oct 2007; revised 27 Dec 2007; accepted 8 Jan 2008; published 16 Jan 2008

0.005%. In fact, the ε_i given by (Γ/ω) (1-ε_r) is not an exactly linear function of Γ, because there is a

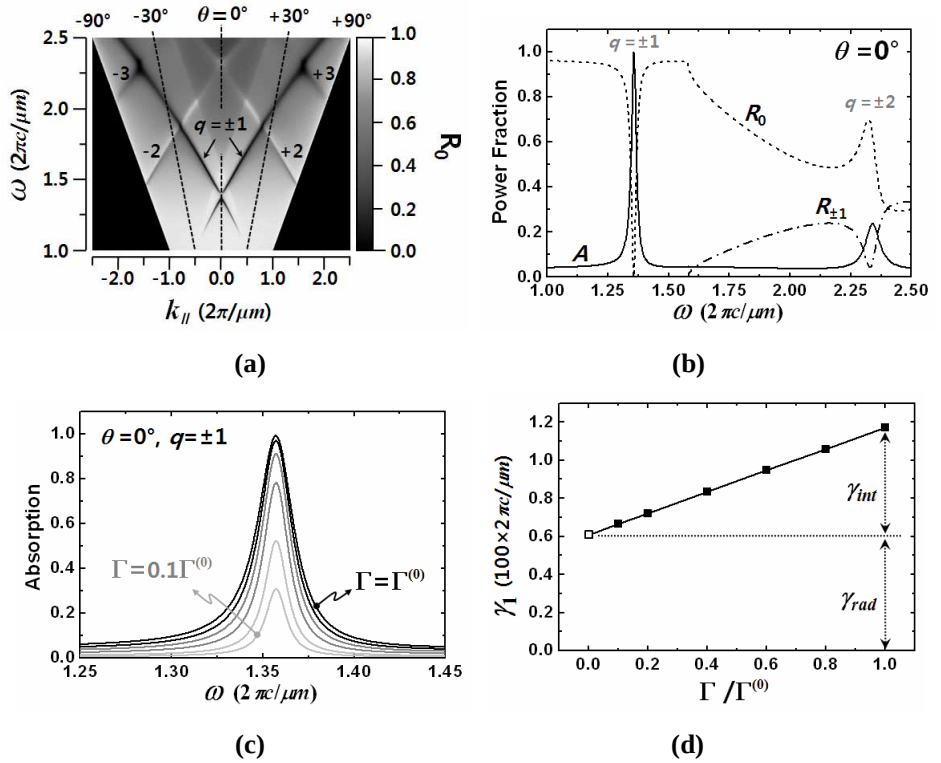

Fig. 2. (a) 0th reflectance as a function of frequency and in-plane wavevector. (b) Absorption, 0th and 1st order reflection for $\theta = 0°$. (c) Magnified absorption spectrum for the 1st order SPR at $\theta = 0°$. A lighter grey curve is corresponding to a smaller collision frequency. (d) Extracted total decay rate as a function of collision frequency by a least-square fitting of (c) to a single-Lorentzian function.

small Γ-dependence in ε_r. But in the limit of $\Gamma/\omega \ll 1$, which is reasonable over a visible frequency range, this ε_r deviation is negligible. In fact, the overall deviation of ε_r is less than 0.6 % in this example case. We can therefore conclude that the total decay rate goes down linearly with the ratio $\Gamma / \Gamma^{(r)}$ over optical frequency range, and that E_{SP} determined by Eq. (4) should be acceptable. In the example structure the resulting extraction efficiency E_{SP} turns out to be E_{SP} =51.9%.

In order to confirm the applicability of the proposed method to a highly confined SPP, further RCWA calculation is made with different grating periods ranging from 50 *nm* to 800 *nm*. All other structural parameters are the same as those used in Fig. 2. Since we are interested in the case where the 1st order diffraction from the SPP gives radiation propagating along the surface normal direction, the method is applied to the 1st order SPRs at $\theta = 0°$. It is also confirmed here how the radiative decay rate and the extraction efficiency vary with frequency and in-plane wavevector. Figure 3(a) shows excellent linearity between γ_1 and Γ for every grating period, where the RCWA results (filled squares) are fitted to straight lines within a maximum deviation of 2×10^{-3}% except for 3.2% maximum deviation at 50 *nm* period of which SPPs resonant at $q = \pm1$ and $q = \pm2$ are close enough to overlap with each other. A sudden change in the line slope between Λ=100 and 50 *nm* is resulted from the fact that the radiative decay rate largely decreases while the rate of internal decay slightly increases. This

#89163 - $15.00 USD Received 29 Oct 2007; revised 27 Dec 2007; accepted 8 Jan 2008; published 16 Jan 2008

behavior is clearly seen in Fig. 3(c) between $k_{//}$=10 and 20. In Fig. 3(b), the positions of [ω_q (0°), $k_{||}$ =2$\pi q/\Lambda$] at q = 1 of the 1st order SPRs are indicated by square symbols. The dark-grey

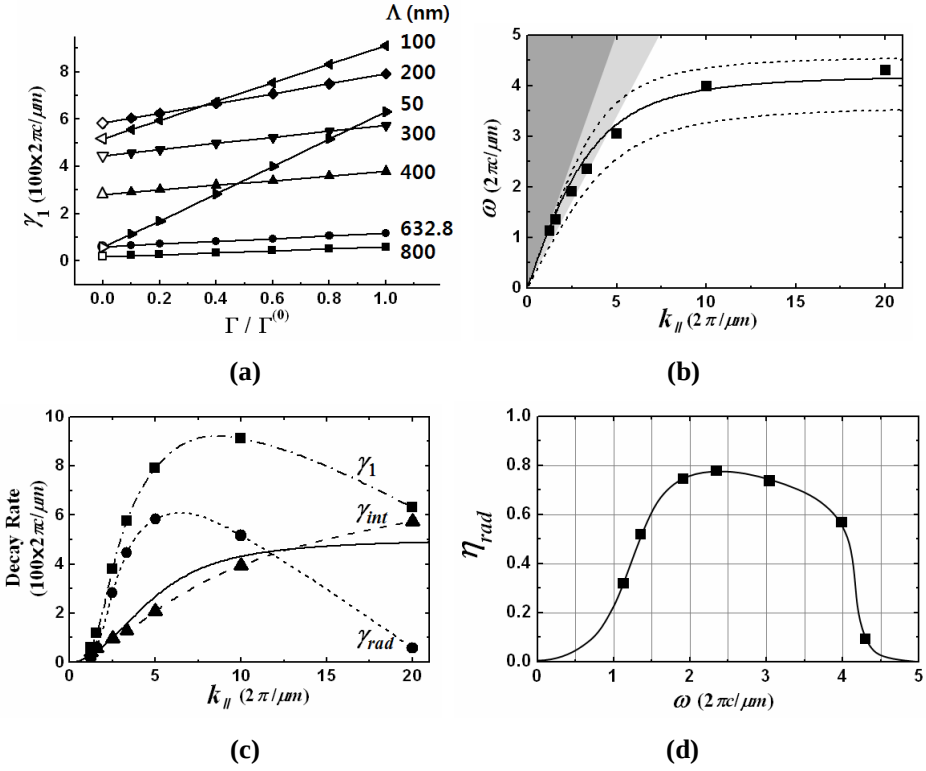

Fig. 3. (a) Total decay rates as functions of collision frequency for different grating periods. They are all extracted from 1st order SPR at θ = 0°. (b) Dispersion curve (square symbols) obtained from 1st order SPR frequencies and primitive Bragg vectors as in-plane wavevectors corresponding to lines indicated in (a). Solid curve tracing the square symbols represents a dispersion of the SPP for the effective dielectric film substituting the grating layer. Upper and lower dotted curve indicates a SPP dispersion for Ag/air and Ag/dielectric interface, respectively. (c) Total (γ_1, squares), radiation (γ_{rad}, circles), and internal (γ_{int}, triangles) decay rates extracted from (a). The solid curve indicates internal decay rate for the effective dielectric film. (d) Resulting extraction efficiency.

and light-grey areas represent the regions where $k_{||} \leq k_0$ and $k_0 \leq k_{||} \leq \varepsilon_g^{1/2} k_0$, respectively, and two dotted curves represent dispersion curves of the SPPs excited on Ag/air (upper curve) and Ag/dielectric (lower) interfaces. The solid curve is the SPP dispersion for an effective homogeneous film instead of the dielectric grating layer. The thickness of the effective dielectric film is the same as the grating depth and its dielectric constant was given by $\varepsilon_{eff}^{-1} = f\varepsilon_g^{-1} + (1-f)\varepsilon_b^{-1}$ [29]. It is apparent that the 1st order SPRs follow on the SPP dispersion of the effective dielectric. In principle, SPP dispersion curves for different periods are not same with each other. But dielectric gratings in our case do not significantly change modal dispersions and corresponding Maxwell-Garnet effective dielectric films are all equivalent. Therefore, the square symbols coincide well with the solid curve, and one can confirm that the absorption peaks analyzed here are originated from to the 1st order SPP excitations. The estimated γ_{int} (triangles), γ_{rad} (circles), and γ_1 (squares) are plotted in Fig. 3(c). γ_{int} (dashed curve connecting triangular symbols) shows approximate consistency with the solid curve indicating the internal decay rate of SPPs for the effective dielectric film. There are slight disagreements between the two curves. For $k_{//}$<13, internal decay rate for the effective dielectric film is

#89163 - $15.00 USD Received 29 Oct 2007; revised 27 Dec 2007; accepted 8 Jan 2008; published 16 Jan 2008

slightly larger than that for the grating. For $k_{//}$>13, however, the situation is reversed. A qualitative explanation can be given in terms of the relative magnitude of radiative decay rate and the Bragg reflection. The electromagnetic mode of a grating system at a SPR can be considered as a set of waves which consists of homogeneous-planewaves running away from the surface and evanescent-waves directly coupling to the SPP. For a low $k_{//}$ limit, the maximum difference is located around $k_{//}$=6. This position corresponds to the largest γ_{rad} as shown in Fig. 3(c). It suggests that, for a larger γ_{rad}, the portion of the energy assigned to the lossless homogeneous-planewave is larger than the other portion attributed to the SPP. Thus, the ohmic loss of the mode should be smaller than that of the SPP on the effective dielectric film which does not include any lossless homogeneous-planewave. For a large $k_{//}$ limit near $k_{//}$=20, on the other hand, γ_{int} for the grating system is larger than that of the effective dielectric film. In this case, γ_{rad} is so small that it gives no remarkable effect any more. The only considerable difference of the grating system from the effective dielectric film is an effect of the Bragg reflection. For the SPRs at θ=0°, standing SPPs are formed because counter-propagating SPPs are identically excited. It is the origin of SPP band gap. At this condition, a mode corresponding to the lower band-edge locates its most energy in the high-index dielectric region and, in fact, it is this mode dealt here. The mode for the grating system, therefore, suffers higher-index dielectric than for the effective dielectric film. As relative penetration depths into a metal and a dielectric roughly satisfy the relation $\delta_{metal}/\delta_{dielectric} = |\varepsilon_{dielectric}/\varepsilon_{metal}|$ [21], the portion of energy in the metal increases for a higher-index dielectric and the mode has more internal losses. Finally, the extraction efficiency E_{SP} shown in Fig. 3(d) is determined as a function of frequency. The frequency around 2.3 shows a maximum efficiency of 78%. Note that the value of 78% is quite large, when compared with the maximum of 50% experimentally obtained in ATR measurement [17]. As we previously discussed on the possibility of underestimation, the ATR measurement must have suffered from leakage radiation back into the high-index prism which degrades the measured extraction efficiency. Also, please note that existence of the maximum in a frequency-dependent E_{SP} is a natural feature of typical SPPs: It results from trade-off between decreasing propagation length and increasing scattering potential of the SPP with increasing frequency. As the frequency approaches the upper cut-off of surface plasmons, the in-plane wavevector of the SPP asymptotically increases far from the light line. This asymptotic behavior gives rise to decreasing group velocity and mode field diameter. As a result, the SPP interact with relatively larger grating for a longer time, leading the mode to scatter more energy into far-field radiation. On the other hand, propagation length of the SPP decreases with frequency. As previously discussed, ohmic loss of the mode is proportional to the ratio of penetration depths, $\delta_{metal}/\delta_{dielectric}=|\varepsilon_{dielectric}/\varepsilon_{metal}|$. Over a lower frequency range, $|\varepsilon_{dielectric}/\varepsilon_{metal}| \ll 1$ because ε_{metal} tends to negative infinity. As the frequency approaches the upper cut-off, ε_{metal} approaches $-\varepsilon_{dielectric}$ and $|\varepsilon_{dielectric}/\varepsilon_{metal}|$ increases up to 1, leading the propagation length, L_{SPP}, to decrease. In addition, it is well known that the propagation length decreases faster than in-plane wavevector because it is linearly proportional to $k_{//}^{-3/2}$ [21]. If we keep the grating period being the same as the SPP wavelength such that $2\pi/\Lambda=k_{//}$, the number of grating grooves that the SPP propagate through is given by $L_{SPP}/\Lambda \propto k_{//}^{-1/2}$. That is, the number of grating grooves decreases to be proportional to $k_{//}^{-1/2}$ with increasing $k_{//}$. Eventually, scattering potential of the SPP for a unit scatterer become larger, but number of bumps with grating grooves become smaller, as the frequency increases. Consequently the maximum extraction efficiency should be given by the balance between them.

#89163 - $15.00 USD Received 29 Oct 2007; revised 27 Dec 2007; accepted 8 Jan 2008; published 16 Jan 2008

3. Correspondence with ATR analysis and applicability to a practical experiment

In order to confirm that E_{SP} determined by Eq. (4) is an upper limit of the conversion energy of a SPP to radiation fields, let's compare the results shown in Fig. 3(d) with those obtained by ATR coupling. Select again, as an example, the 1st order SPR for Λ = 400 *nm* represented

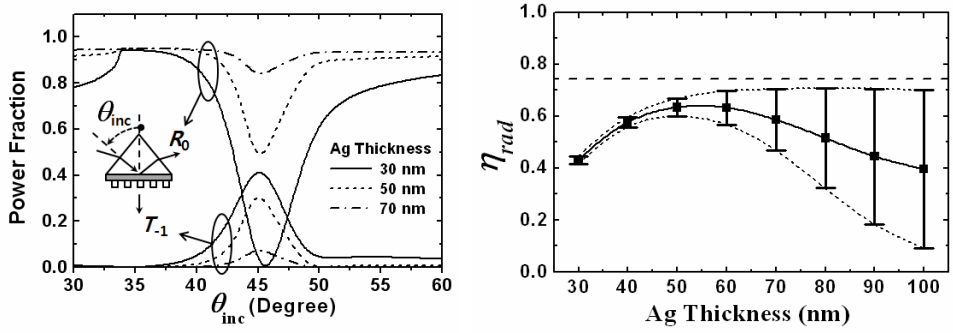

Fig. 4. (a) Calculated ATR spectrum for several Ag thicknesses. The SPR is corresponding to 1st order SPR for Λ = 400 *nm* in Fig. 3. (b) Extraction efficiency obtained by ATR method. Dashed line represents the reference value of efficiency taken from Fig. 3 (d) at ω = 1.91.

by the upward-triangular symbol (▲) in Fig. 3(a). The proposed method exhibits E_{SP} = 74.5% in this example case when ω_1 = 1.91 (λ_0 = 524 *nm*) and k_{SP} = 2.5. In ATR-coupling calculation, we replace the infinite metal shown in Fig. 1 by an Ag film with a finite thickness ranging from 30 *nm* to 100 *nm* attached on a prism with refractive index of 1.8, while the background dielectric (air) and the grating structure keep constant. The ATR absorption minimum corresponding to the SPR is at θ_{inc} = 46.7° for the three different thicknesses as shown in Fig. 4(a). Both of the absorbed power fraction in R_0 and the transmitted one in the 1st order SPR, T_{-1}, are simultaneously reduced after Ag thickness of 30 *nm* where R_0 and T_{-1} are extreme. E_{SP} for the ATR coupling method is estimated by the same procedure as ref. 15 and 16, and the results at different Ag thicknesses are depicted in Fig. 4(b). The dashed horizontal line represents E_{SP}=74.5% as a reference. The error bars indicate the upper and lower efficiencies respectively obtained by the relations of T_{-1}/(R_0'-R_0) and T_{-1}/(1-R_0) as explained earlier, and the square symbols represent their averages. R_0', an estimation of the reflectance when there is no SPR, is determined by a cubic interpolation of the R_0 curve after eliminating its absorption deep. The upper efficiencies, which are quite different from the lowers, approach the reference value of 74.5% as the Ag film become thicker. The leakage radiation from the SPP towards the prism becomes negligible after about 60 *nm* in the Ag thickness, thus a thinner metal case must considerably suffer from a larger leakage radiation and subsequently produces an underestimation of E_{SP}. Consequently it is evident that the reference value determined by the proposed method should be an upper limit of E_{SP}.

It is worth discussing again whether the linear relationship between the total decay rate of SPPs and ε_i in Eq. (2) can be acceptable in most use of noble metals, and whether collision frequency or temperature dependence of the total decay rates can be linearly extrapolated as a function of Γ down to Γ / $\Gamma^{(r)}$ = 0 starting from $\varepsilon_i^{(r)}$ measured at a higher temperature. It is well known that the electron relaxation time for DC electric current is given by [30]

$$\frac{1}{\tau_{DC}} = \frac{4}{\tau_0}\left(\frac{T}{T_D}\right)^5 \int_0^{T_D/T} \frac{Z^5}{(e^Z - 1)(1 - e^{-Z})} dZ \,, \qquad (5)$$

#89163 - $15.00 USD Received 29 Oct 2007; revised 27 Dec 2007; accepted 8 Jan 2008; published 16 Jan 2008

where T_D is Debye temperature (about 225K for Ag), and τ_0 is a characteristic relaxation time independent on frequency for isotropic collision process. This expression for a DC collision frequency gives two limiting dependences such that $\Gamma_{DC} = 1/\tau_{DC} \simeq 2T/(3\tau_0 T_D)$ for $T \gg T_D$ and $\Gamma_{DC} \simeq 0$ for $T \ll T_D$. As temperature decreases, the collision frequency linearly decreases and finally vanishes along $(T/T_D)^5$.

In optical frequency range, however, a special care must be taken to account for the experimental validity because the collision frequency of electrons does not tend to complete zero at low temperature limit. This is attributed to a phonon assisted light absorption which cannot be ignored for $\hbar\omega \gg k_B T$. A theoretical prediction considering the 2nd order perturbation for electron transition and the rate of electromagnetic energy transfer in an optical frequency gives the temperature dependence of the collision frequency over an optical frequency range such that [31],

$$\Gamma_{opt} = \frac{1}{\tau_{opt}} = \frac{1}{\tau_0}\left[\frac{2}{5} + 4\left(\frac{T}{T_D}\right)^5 \int_0^{T_D/T} \frac{Z^4}{(e^Z - 1)} dZ\right]. \quad (6)$$

The temperature dependence of Γ_{opt} is obviously different from Γ_{DC}, especially for low temperature limit. The temperature-independent term of '2/5' in the bracket of Eq. (6) is corresponding to the phonon assisted light absorption, and thus gives residual non-vanishing relaxation at 0K. It results in sustaining non-zero collision frequency, $\Gamma_{opt} \simeq 2/(5\tau_0)$, at low temperature limit and degradation of linearity of the temperature dependence at a high temperature range. The corresponding residual imaginary dielectric constant of a metal at zero temperature is given by $\varepsilon_i = 2(1-\varepsilon_r)/(5\omega\tau_0)$. Tillin *et al* had measured this residual value of a flat Ag film by about 29% of the value at room temperature for 632.8 *nm* wavelength by fitting ATR absorption spectrum [31]. Therefore, in optical frequency range it is not possible to completely eliminate the ohmic decay of SPPs by lowering temperature. It does not mean, however, that our proposed method to estimate E_{SP} by changing ε_i is not effective any more. Though the phonon assisted absorption will make the temperature dependence of the decay rate of a grating-induced SPPs a little bit complex, the smooth extrapolation to $\varepsilon_i = 0$ in Eq. (3) will still be valid under the reasonable assumption that the residual ε_i are not so high relative to the values at room temperature. According to ref. 31, for example, ε_i (77K)/ ε_i (300K) $\doteq$ 0.36 in an annealed Ag film at 632.8 *nm* wavelength. It means that the Ag film in a liquid-nitrogen flow cryostat reveals a 64% reduction in an internal decay rate of the SPP. The non-zero residual does not lead to any abrupt change in temperature dependence of the SPP decay rates between room temperature and lower temperature limit. The temperature dependence should be a moderate function for a certain temperature range where there is no phase transition. Therefore, the linear relationship between SPP decay rate and ε_i can be acceptable in most use of noble metals and the temperature dependence of SPP decay rates can be straightened into a linear function of ε_i in such a temperature range.

4. Conclusion

We have proposed a unique method to determine the upper limit of radiative extraction efficiency from SPPs, E_{SP}, based on measurement of the spectral bandwidth of a grating-induced SPR absorption spectrum as a function of metal dielectric constant. When $|\varepsilon_r| \gg |\varepsilon_i|$, we have found from RCWA calculations that the SPP decay rate has an extremely linear relationship with the collision frequency Γ of metal and that the upper limit of E_{SP} can be

#89163 - $15.00 USD Received 29 Oct 2007; revised 27 Dec 2007; accepted 8 Jan 2008; published 16 Jan 2008

determined by the asymptotic extrapolation of a total decay rate of the SPP at Γ=0. The proposed method is inherently free from the ambiguity and underestimation difficulties pertaining to other methods such as ATR-coupling measurements. It can also be applied to highly confined SPPs excited in a SPP-mediated LED where E_{SP} becomes a dominant factor in determining the total external efficiency of LEDs. Especially for the case where the SE rate is greatly enhanced by SPP excitation, this method would excellently figure out the maximum realizable external efficiency since it converges on E_{SP} in this extreme case.

Acknowledgements

This work was supported by grant No. R01-2005-000-10276-0(2007) from the Basic Research Program of the Korea Science & Engineering Foundation.

#89163 - $15.00 USD Received 29 Oct 2007; revised 27 Dec 2007; accepted 8 Jan 2008; published 16 Jan 2008