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Can lasing at visible wavelengths be achieved using the low-loss long-range surface plasmon-polariton mode?

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Abstract. The use of emissive dye molecules as a gain medium in order to excite the low-loss long-range surface plasmon-polariton (SPP) mode on a thin silver film has previously been suggested as a method of achieving SPP lasing at visible wavelengths. Here, we consider the gain lost to the short-range SPP, an aspect that has not been investigated before and which may preclude the possibility of lasing. We show by experiments that gain will indeed be lost to the short-range mode. From a computational study, we find that despite gain lost to the short-range mode, SPP based lasing may still be possible using the long-range mode, although we identify a number of requirements that might make lasing difficult to achieve in practice.

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Contents

1. Introduction

Recently the suggestion was made that a laser based on the surface plasmon-polariton (SPP) mode and operating in the visible part of the spectrum might be possible [1, 2]. Such a demonstration would be of great interest to those working on developing new components for photonics based on SPPs [3, 4], part of a field now referred to as plasmonics. SPP modes have been demonstrated to be capable of acting as waveguides over centimetre scale distances [5], the propagation length ultimately being limited by absorption losses in the metal. To show that by adding a gain medium such losses could be overturned and lasing made possible might well act as a spur to much new activity. Lasing using SPP modes has been achieved at infrared wavelengths from 3.4 μm [6] to beyond 90 μm [7], in quantum cascade lasers. Absorption losses in metals at visible wavelengths are much greater than those at infrared wavelengths so that a higher-gain medium would be required; even then the gain would not appear to be sufficient to achieve lasing using the usual SPP mode. To address this Okamoto *et al* [2] recently proposed using a lower loss SPP mode, the so-called long-range SPP (LRSPP) mode of a symmetrically clad thin metal film [8]. However, such metal films support two coupled SPP modes, one long-range, the other short-range. The calculations of Okamoto *et al* [2] did not take account of the gain that would be lost to this second (short-range) SPP mode. Below we present experimental data which show that both modes need to be taken into account, and perform calculations to see whether, even in the presence of this mode, lasing is still possible. We find that lasing might in principle be possible, but that fabrication requirements are likely to be a challenge.

SPPs are electromagnetic modes that are supported by a metal–dielectric interface [9]. They are waves that are trapped at the surface and which decay exponentially into both the metal and dielectric media. A metal film will support a SPP mode on each surface. If the metal is thin enough then the fields associated with these two modes may overlap in the metal. Further, if the media bounding the metal have the same refractive index then, for any given frequency, the two SPP modes will have the same in-plane wavevector and in this case will couple together so as to produce two coupled non-degenerate modes. One of these coupled modes has a symmetric

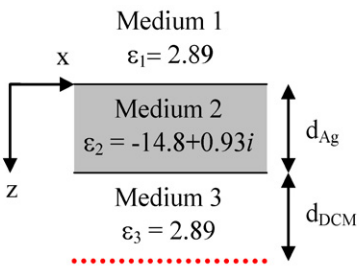

Figure 1. Thin metal film clad with dielectric material. The separation between the metal film and the dipole emitters, d_{DCM}, is used for simulation purposes. The thickness of the silver layer is indicated by d_{Ag}. The wavelength used is 620 nm, the peak emission wavelength of the dye, DCM.

charge distribution and thus has fields that tend to be screened from the metal, so reducing the effect of absorption on the mode, relative to the single interface mode. This mode therefore has a longer propagation length than the single interface mode and is often known as the LRSPP. The other coupled mode has an anti-symmetric charge distribution with fields that are concentrated more in the metal than the single interface mode—this mode is often referred to as the short-range SPP (SRSPP).

To make a SPP laser, a gain medium needs to be placed close to one of the metal surfaces; specifically, it needs to be within the field distribution of the SPP modes. Both LRSPPs and SRSPPs will act as power sinks for the emitters that comprise the gain material. If a large fraction of the total power emitted by the gain material couples to the SRSPP, then the large loss within the system could negate the possibility of lasing. Here, we theoretically model the structures studied by Okamoto *et al* [2] and calculate the fraction of the power that is coupled to both the LRSPP and the SRSPP.

2. Investigating the power coupled to the long- and short-range modes of a symmetric structure: theory

Okamoto's proposed structure consisted of a substrate with a refractive index of $n = 1.7$, upon which was deposited a thin silver film. To enable the structure to support coupled modes, the silver was coated with a dielectric layer which also had a refractive index of 1.7. The material chosen was 4-dicyanomethylene-2-methyl-6-*p*-dimethyl-aminostyryl-4H-pyran (DCM)-doped tris-(8-hydroxyquinoline) aluminium (Alq_3) which has a refractive index of 1.7 at the peak emission wavelength of DCM, 620 nm; the geometry of this sample is shown in figure 1. As noted above, if the silver film is thin enough the evanescent fields of the SPP modes on either side of the metal may overlap to form a pair of coupled modes. The authors calculate that for samples having a silver film less than 60 nm thick, the gain will exceed the loss of the LRSPP mode, thus allowing such a laser to reach threshold.

We begin our investigation by calculating the fraction of power coupled from the emissive molecules to each SPP mode, and thus calculate the gain and loss coefficients of both modes. Reference [10] gives extensive background concerning details of the model used, a model based on a classical antenna approach. The model works by representing the dipole field of an emitter

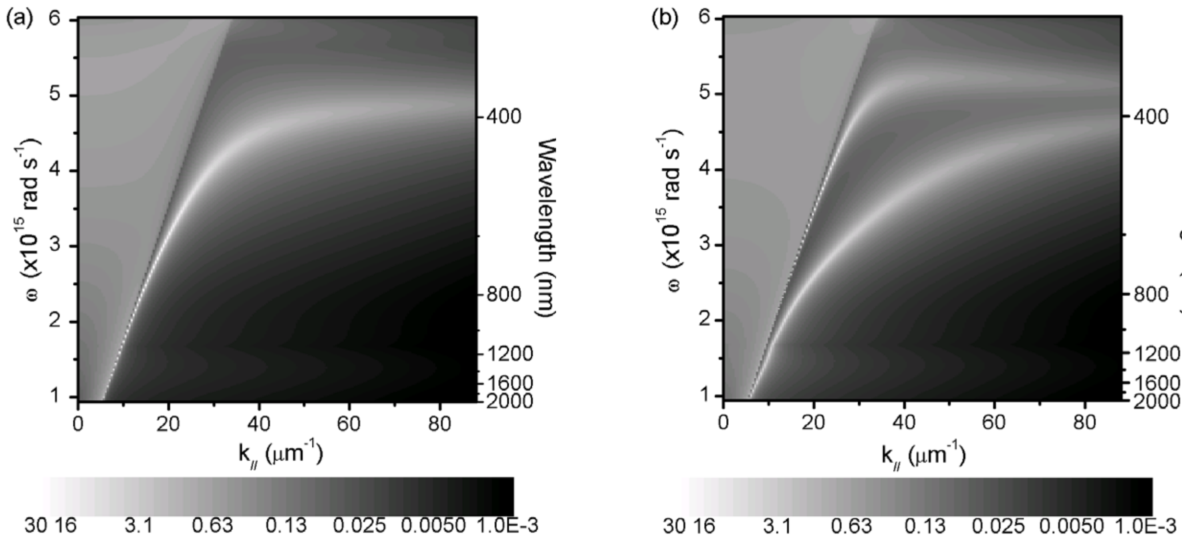

Figure 2. Power (arbitrary units) lost by an isotropic dipole as function of in-plane wavevector ($k_{\|}$), with intensity shown on a logarithmic scale; white regions indicate strong coupling of source to mode. The silver is (a) 140 nm thick; (b) 20 nm thick. The light line is that of the bounding material which has a refractive index of 1.7. The separation between the emissive dipole and the metal is 20 nm.

(such as an excited dye molecule) as a sum of plane-waves with different in-plane wavevectors (the wavevector in the plane of the interfaces). The power dissipated from the dipole is calculated as a function of the in-plane wavevector at a specific emission wavelength, from which a power dissipation spectrum can be constructed [11]–[14]. The power lost by an emissive dipole as a function of the in-plane wavevector was simulated for two samples having the structure indicated in figure 1, one having a 140 nm thick silver film, the other having a 20 nm thick silver film; the results are shown in figure 2.

Figure 2(a) shows the dispersion (angular frequency as a function of in-plane wavevector) of the single interface SPP present on a silver film 140 nm thick. The data are shown on a logarithmic scale, with the white regions denoting strong coupling between the emitter and the mode. Figure 2(b) shows how, when the silver film is only 20 nm thick, the modes associated with the two metal surfaces interact to produce two coupled modes.

The dispersion curves have been calculated with the emissive source positioned within one of the dielectric layers, 20 nm from the silver film. The position of the source does not affect the dispersion of the SPP modes, however it does alter the strength with which the dipole source couples to the two modes. The permittivity of the silver has been calculated from polynomials fit to the data collated by Palik [15].

To better understand the nature of the coupled modes seen in figure 2, field profile calculations were carried out using a procedure similar to that adopted by Kovacs [16] and based on solving Maxwell's equations in the presence of appropriate boundary conditions. The simulations assume that there is no incident wave; data are normalized such that the amplitude of the electric field transmitted from medium 1 to medium 2 is initially set to unity at the boundary; reflected fields from subsequent boundaries alter this amplitude. The fields at any given point

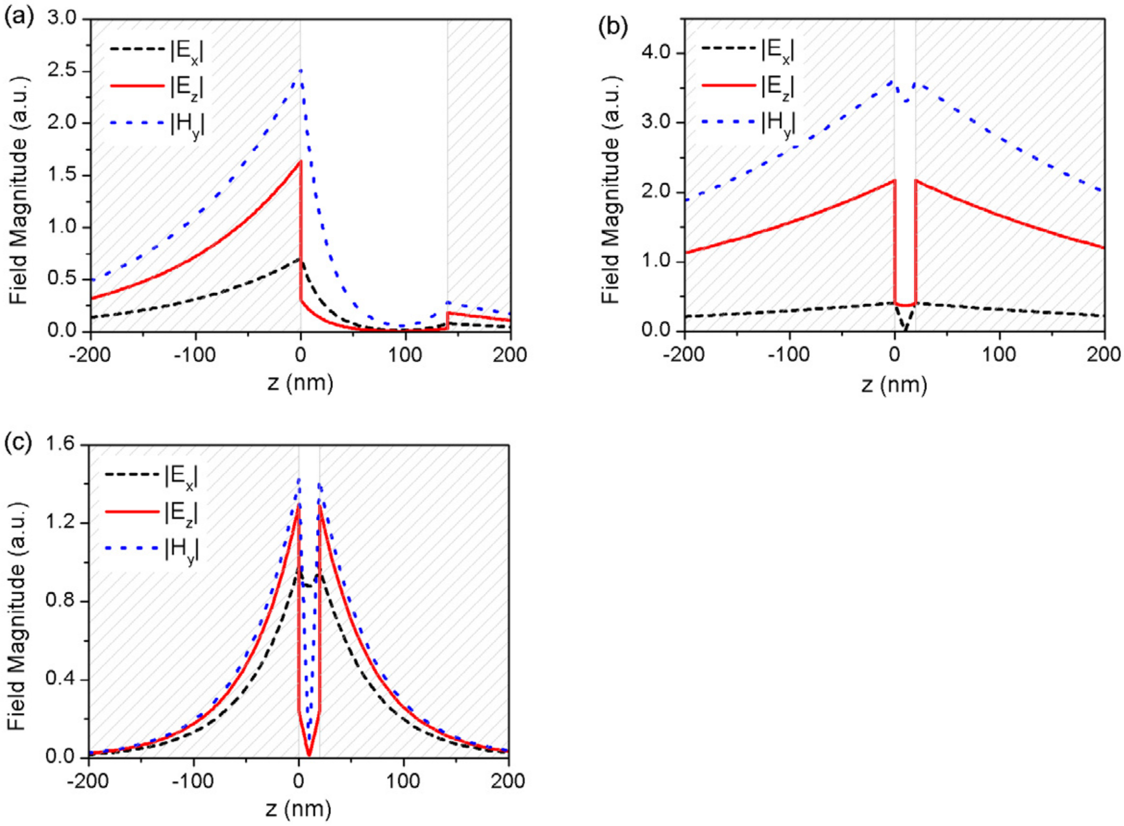

Figure 3. The magnitudes of the field components E_x, E_z and H_y as a function of distance through the sample, z. The fields shown are for (a) the single SPP mode on 140 nm silver; (b) the LRSPP across 20 nm silver and (c) the SRSPP across 20 nm silver. The shaded regions indicate the bounding media with $n = 1.7$, and the unshaded regions indicate the silver.

are calculated from the net contribution due to all reflections. The **H** fields are scaled by c, so as to allow E and H field components to be plotted on the same scales.

The nature of single- and coupled-SPP modes can be understood from figure 3, which shows the field profiles for (a) the single interface SPP present on the 140 nm thick silver film, (b) the LRSPP across the 20 nm film and (c) the SRSPP across the 20 nm thick film.

The fields associated with the single SPP are localized on one surface of the metal; they are strongest at the metal–dielectric interface and decay exponentially into both the metal and the dielectric. In contrast, the fields of the coupled modes are not localized on one particular interface but instead span the metal film; the fields of the LRSPP are weak within the metal and decay slowly into the dielectric; the fields of the SRSPP are stronger in the metal, and decay more quickly into the dielectric. The data in figures 2 and 3 show clearly that when the silver film is thin, an emissive dipole near one of the metal surfaces will couple to both the LRSPP and SRSPP modes.

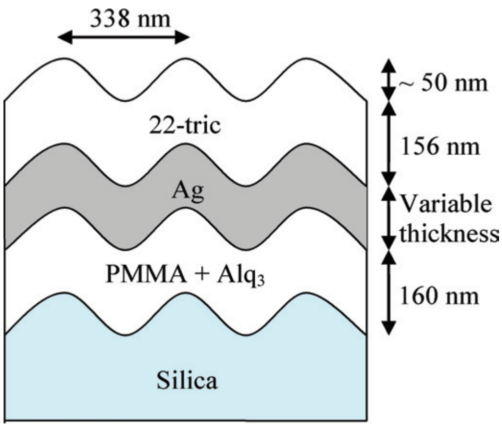

Figure 4. Sample structure. Numerous samples were produced, each with a different thickness of silver. The thickness of the 22-tric layer was calculated to give the SPPs on either surface of the silver the same nominal wavevector.

3. Experimental demonstration that both long- and short-range modes are excited by potential gain media

To show that both modes act as important decay routes for adjacent excited molecules, we studied the coupling of power to the two SPP modes supported by a variant structure [17], one that consisted of a silica substrate with a grating profile etched into it with a pitch of 338 ± 1 nm and an amplitude of ~50 nm. The grating was necessary to enable the SPPs to couple to light so that an experimental dispersion diagram could be obtained. To provide an emissive layer, the substrate was covered by spin-coating with 160 nm poly(methyl methacrylate) (PMMA) doped with Alq_3 (3% by weight). Subsequently silver was deposited by thermal evaporation under vacuum so as to yield nine regions with silver thicknesses ranging from 20 to 90 nm. Finally, to provide the symmetric structure needed for coupled surface plasmon modes, a multi-molecular organic thin film 156 nm thick, made of 22 tricosenoic acid monolayers (22-tric) was deposited by the Langmuir–Blodgett technique. The thicknesses of the two organic media (PMMA ($n = 1.46$) and 22-tric ($n = 1.58$)) were chosen to give the SPPs on either surface of the silver the same nominal wavevector, thus ensuring that the surface plasmon modes on the two silver surfaces would couple. The structure of this sample is shown in figure 4.

To examine the modes of this sample, the Alq_3 was optically pumped through the substrate by a 410 nm diode laser. The resulting photoluminescence (PL) was collected over a range of angles. Figure 5 shows the PL obtained from samples with silver film thicknesses of 20 and 90 nm (figures 5(a) and (b) respectively); the data have been plotted as a function of frequency and in-plane wavevector so that they form dispersion diagrams. Two scattered SPP modes can be seen in each case; the modes clearly move closer together as the silver is made thicker, confirming that the modes observed are coupled. The high frequency mode is the LRSPP, and the low-frequency mode is the SRSPP [8]. The data clearly show that emissive molecules adjacent to a thin metal film will couple to both the LRSPPs and SRSPPs. This means that the gain available to the LRSPP is reduced as a consequence of power lost to the SRSPP mode. The data presented here do not allow us to quantify how much power is lost to the two modes, since the intensity associated with each mode in this experiment depends not just on the power lost to that mode but also on

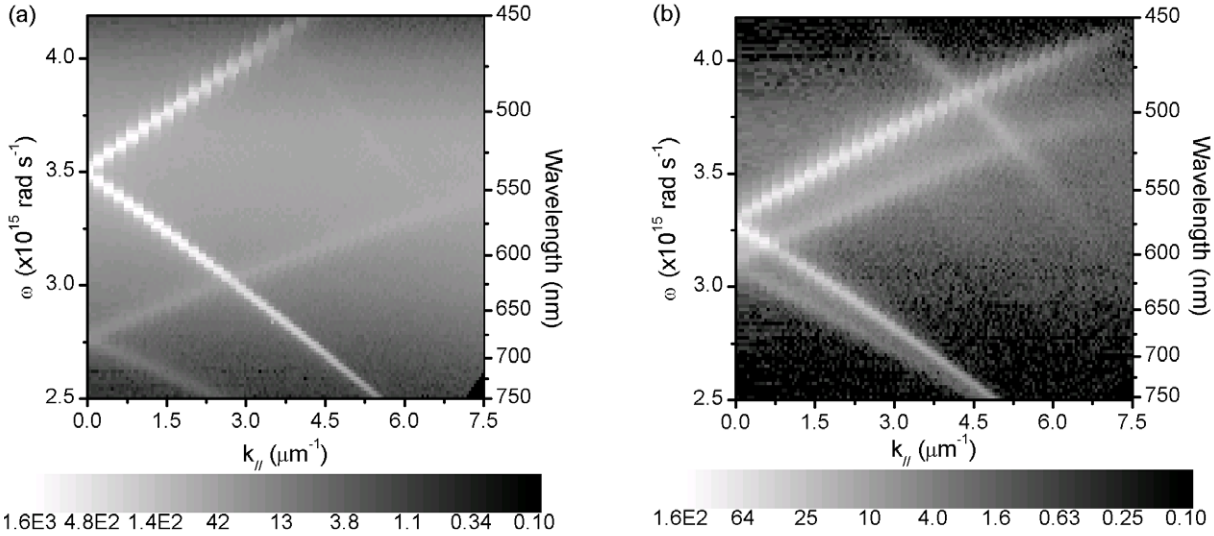

Figure 5. PL collected from samples with a silver thickness of (a) 20 nm and (b) 90 nm. The samples were pumped optically through the substrate and the PL collected from the 22-tric side. The scattered long- and short-range modes can be seen in each case; white regions indicate high intensity. The higher wavevector mode that is present in both cases is a TM-polarized guided mode within the PMMA layer.

the effectiveness with which that mode may be coupled to light. As we have shown elsewhere, the latter process can be a very subtle one [18].

4. Theoretical modelling

Having established that the presence of the short-range mode is something we need to take into account when thinking of using the LRSPP mode for lasing there are now two important questions to be considered. Firstly, how does the loss of power to the SRSPP affect the possible gain in the system. Secondly, is there a way to minimize this loss and thus optimize the gain available to the long-range mode?

4.1. Calculating the loss coefficient of a mode

To find the loss coefficient of SPP modes, power dissipation spectra, i.e. graphs showing the power dissipated from an emissive dipole as a function of in-plane wavevector ($k_\parallel$), can be employed. SPP modes are observed as peaks in the power dissipation spectrum, at wavevectors greater than those possible for photons in the adjoining media, i.e. beyond the light-line. The width of a SPP mode is proportional to the loss coefficient of the mode. The power-dissipation spectra are calculated at a single frequency in the same manner as the dispersion curves shown in figure 3. The data in figure 6 show the power dissipated by an emitter as a function of in-plane wavevector from an emissive dipole, normalized with respect to the free space wavevector, k_0. Here, we have assumed that the dipole moment associated with the emitter samples all directions

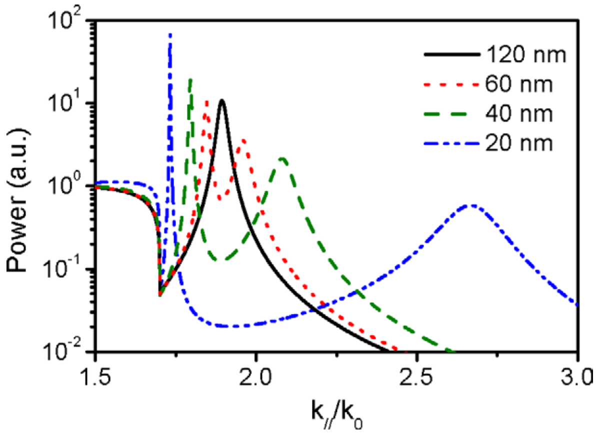

Figure 6. Power dissipation versus normalized in-plane wavevector for a source near silver films of various thicknesses clad in a medium with $n = 1.7$. The emissive dipole is situated within one of the dielectric layers at a distance of 90 nm from the silver ($\varepsilon_{\mathrm{Ag}} = -14.8 + 0.93\mathrm{i}$ at the wavelength 620 nm).

in space on a timescale faster than the excited state lifetime. We refer to this as the isotropic orientation, a situation common to small molecule emissive species, such as many dye molecules. Furthermore, for the purposes of our modelling, we have taken the quantum efficiency of the emitters to be unity. All simulations in subsections 4.1–4.3 assume that the sample has the planar structure indicated in figure 1, with $\varepsilon_1 = \varepsilon_3 = 2.89$. Subsection 4.4 investigates the effect of varying ε_1 slightly to produce an asymmetric structure.

The data shown in figure 6 are all simulated at the peak emission wavelength of the dye proposed by Okamoto *et al* [2], DCM (620 nm). The dipole is situated within a dielectric of $n = 1.7$, at a distance of 90 nm from the near surface of a silver film, and calculations were performed for silver films of varying thicknesses. Power emitted with $k_{\parallel}/k_0 < 1.7$ is emitted radiatively into the dielectric bounding media. The structure having a silver thickness of 120 nm shows only one SPP peak, whereas the thinner films show two peaks, a sharp LRSPP and, at higher wavevectors, a broader SRSPP. The two modes move closer together as the silver becomes thicker because the fields associated with modes overlap less and there is therefore less interaction. The data for the 120 nm silver film shows no interaction between the modes.

As noted above, the attenuation associated with a SPP mode may be calculated from the width of the mode. By employing the data shown in figure 6, and similar data calculated for structures having different thicknesses of silver, the attenuation of each mode as a function of silver film thickness could be determined (figure 7). The loss of the LRSPP mode decreases dramatically as the silver is made thinner, but the loss associated with the SRSPP mode increases.

4.2. Fraction of power coupled to each SPP mode

Our next step is to determine the fraction of power coupled to each SPP mode. By integrating the power under the various peaks in the curves shown in figure 6, and similar curves for other silver film thicknesses, the fraction of the total power dissipated by the molecules that is coupled to each mode can be calculated. Figure 8 shows the fractional power coupled to each mode as a function of the silver film thickness. This has been calculated with the dipole situated 20 nm and

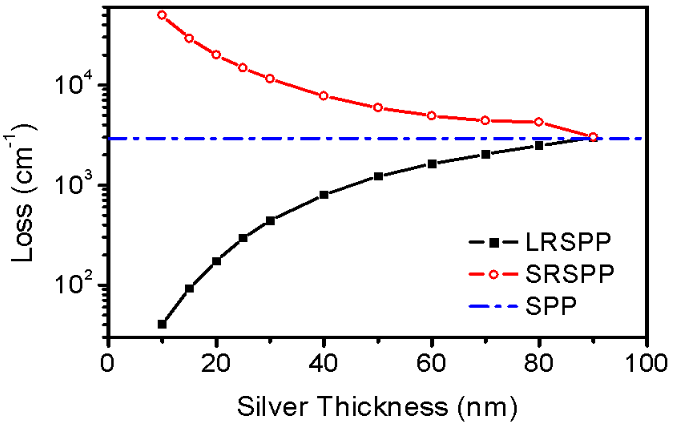

Figure 7. The calculated loss coefficients of the LRSPP and SRSPP modes as a function of silver film thickness. The emissive dipole is situated 90 nm from the silver within one of the dielectric layers ($n = 1.7$). The loss coefficient of a single SPP mode on a 250 nm thick silver film clad in the same dielectric is shown for comparison.

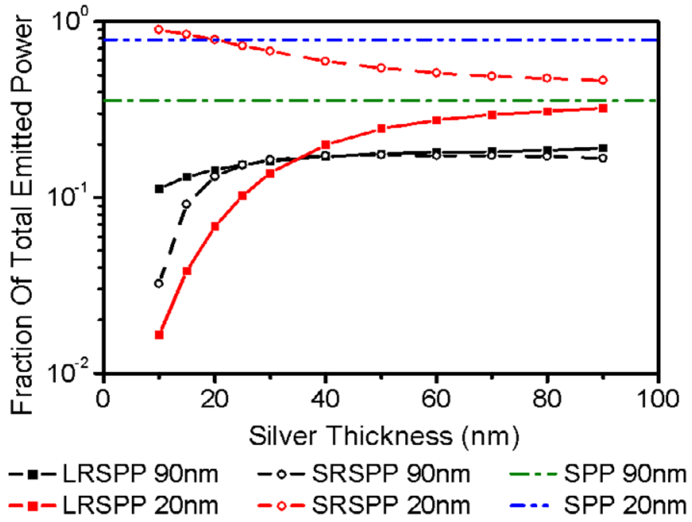

Figure 8. The fractional power emitted to the LRSPP and SRSPP as a function of silver film thickness, for samples with the emissive dipole situated 20 or 90 nm from the silver film. The fraction of power emitted to the single SPP on a 250 nm thick silver film is shown for comparison in each case.

then 90 nm from the silver. Previous research has shown that the separation between the dipole and the silver film has a large effect on the amount of power that couples to each mode [19]. The reason for this can be seen in the field profiles shown in figure 4; the field components of the SRSPP decay very quickly with distance from the film, thus a dipole at 90 nm will experience much weaker fields than a dipole at 20 nm, and therefore couple less power into the SRSPP mode. The fields of the LRSPP mode extend much further into the dielectric so the magnitude

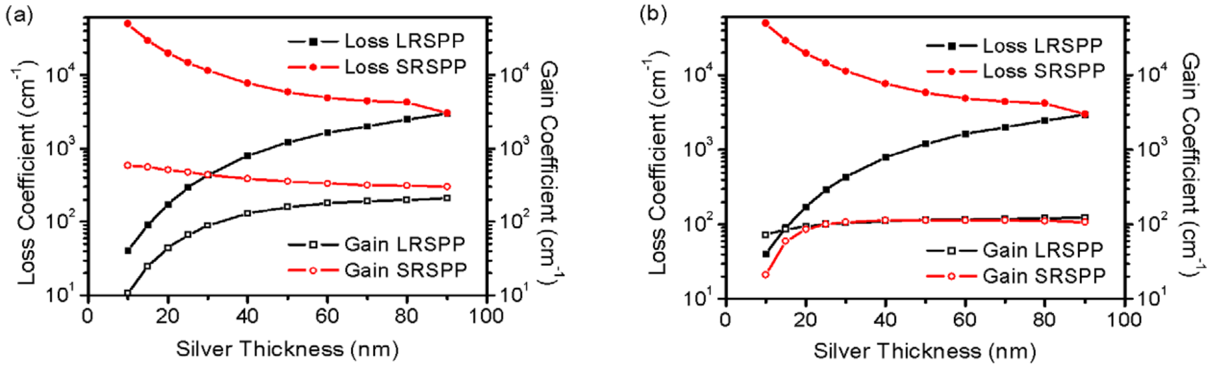

Figure 9. The loss and gain coefficients of the LRSPP and SRSPP modes as a function of silver film thickness for samples with the emissive dipoles situated (a) 20 nm and (b) 90 nm from the silver film.

decreases only slightly as the dipole is moved from 20 to 90 nm away. The amount of power entering the LRSPP mode will therefore decrease only slightly over this distance. The loss of power from the SRSPP mode means that the fraction of the total power emitted from the dipole that is available for gain within the LRSPP will increase.

4.3. Gain coefficients of SPP modes

The dye, DCM, is reported to have a maximum gain of ~650 cm^{-1} [2]. The gain of each coupled mode will therefore be a fraction of this total gain, proportional to the amount of power coupled to each mode. If, for instance, 20% of the total emission couples to the LRSPP, the maximum gain available to this mode would be 20% of the total gain available, or 130 cm^{-1}. To calculate the net gain available to each mode, the loss coefficient of the mode must be subtracted from this gain coefficient.

The gain coefficient of each mode was calculated in the manner described above as a function of silver film thickness. The results are plotted, together with the loss coefficient of each mode, in figure 9. When the separation between the dipole and the silver is 20 nm (figure 9(a)), the loss of each mode is always greater than the gain available to that mode, so that lasing is not possible. However, when the separation is increased to 90 nm (figure 9(b)), thus greatly increasing the relative amount of power coupled to the LRSPP mode, the gain available to the LRSPP is greater than the loss associated with the mode, providing the silver thickness is less than 15 nm. This threshold thickness is much lower than the 60 nm threshold suggested in the original paper by Okamoto *et al* [2], and is due to the fact that here we have taken account of the reduction in gain available to the LRSPP mode as a consequence of the presence of the SRSPP. The data in figure 9(b) indicate that thinner silver films lead to more net gain being available to the LRSPP mode. However, silver films thinner than ~15 nm typically have greater absorption [14], so achieving the low-loss thin silver films needed here is likely to be a challenge.

Since the separation between the dipole and the silver has such a large effect on the possible gain in the system (figure 9), power dissipation data were computed for a wide range of molecule–metal separations, with a 10 nm thick silver film in each case. The data calculated with separations

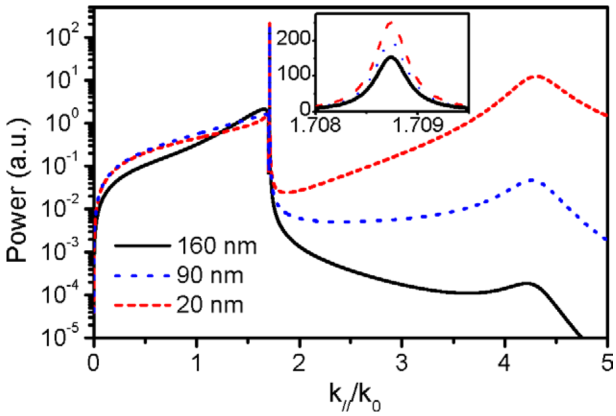

Figure 10. The power dissipated from an emissive source situated at various distances from a 10 nm thick silver film, as a function of normalized in-plane wavevector. The inset shows the LRSPP peak on a linear scale.

of 20, 90 and 160 nm are shown in figure 10. As previously mentioned, the wavevectors of the SPP peaks are unchanged when the molecule–metal separation changes. However, the amount of power coupled to the LRSPP decreases by a factor of two as the separation increases from 20 to 160 nm (inset). In contrast, over the same separation range the amount of power coupled to the SRSPP ($k_\parallel/k_0 \sim 4.2$) decreases by over five orders of magnitude. Hence, as the separation is increased from 20 to 90 nm the fraction of power coupled to the LRSPP mode increases from 1.7 to 11%, thus greatly increasing the possible gain; the fraction of power coupled to the LRSPP subsequently falls as the dye–silver separation is increased further, to 10.1% when the separation is 160 nm.

To explore this further, the net gain as a function of the dipole–silver separation was calculated by subtracting the loss coefficient from the gain coefficient, with the silver film either 10 or 15 nm thick (figure 11). From figure 11, it may be seen that samples containing 15 nm silver will never achieve lasing threshold (indicated by the dashed line), however, samples with 10 nm silver can show a gain when the separation is between 45 and 250 nm.

4.4. Asymmetric structures

A further possible method of decreasing the loss associated with the long-range mode was put forward in a paper by Wendler and Haupt [20]. They showed that introducing a slight asymmetry into the system by varying the permittivity, ε_1, of one of the dielectric layers can allow the propagation length of the LRSPP mode to increase by up to three orders of magnitude. Upon investigation however, by computing the power dissipated as a function of in-plane wavevector we show that the long-range mode actually splits into a properly guided mode very close to the light line, and a leaky mode having a wavevector just inside the light line [21].

The various modes can be seen in the power dissipation spectra shown in figure 12; the data shown is for a sample with $\varepsilon_1 = 2.884$ ($n = 1.68$). The sample has the optimized geometry described above; 10 nm of silver and a dye–silver separation of 90 nm. Comparing these data to those shown in figure 10, it can be seen that the position of the SRSPP remains unchanged, whereas the LRSPP has split. The inset in figure 12 shows the leaky mode ($1.68 < k_\parallel/k_0 < 1.7$) and the sharp guided LRSPP mode just on the light line ($k_\parallel/k_0 = 1.7$). When the difference

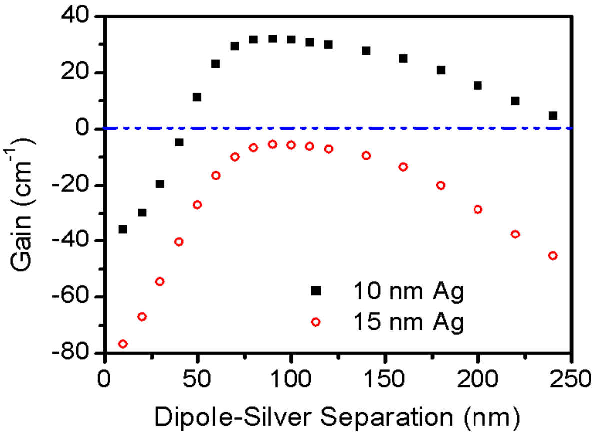

Figure 11. The gain from samples containing 10 or 15 nm silver, as a function of the separation between the emissive dipole and the silver. The dashed line indicates the lasing threshold.

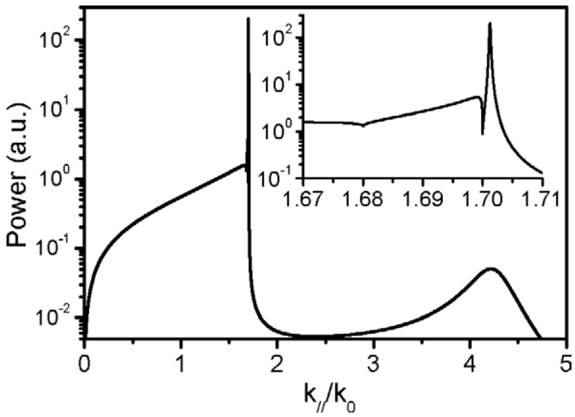

Figure 12. The power dissipated as a function of in-plane wavevector for an asymmetric-permittivity structure with $\varepsilon_1 = 2.8224$. The inset shows the leaky mode and the guided LRSPP on a larger scale.

between ε_1 and ε_3 becomes greater, the dipoles couple less power into the guided LRSPP mode until all of the power is coupled simply to the leaky mode and the SRSPP; the leaky mode is unsuitable for lasing purposes.

The loss of the guided LRSPP mode and the fractional power dissipated to this mode was calculated, and the total possible gain available to the guided LRSPP mode was calculated in the same manner as that described above. We find that in the asymmetric system the loss for the guided mode can be less than that of the LRSPP of the symmetric system. However, in the asymmetric system, the gain is also reduced as some power is lost to the leaky mode. As a result the overall gain coefficient of the guided long-range mode only varies slightly from the symmetric case. The total possible gain of the guided LR mode as a function of the permittivity of the dielectric layer is shown as the solid line in figure 13. The dashed line indicates the symmetric case, $\varepsilon_1 = \varepsilon_3 = 2.89$. The gain is a maximum for $\varepsilon_1 = 2.876$, however, the actual increase in gain is very slight, less than 3%, and fabrication of such a sensitive structure may require more effort

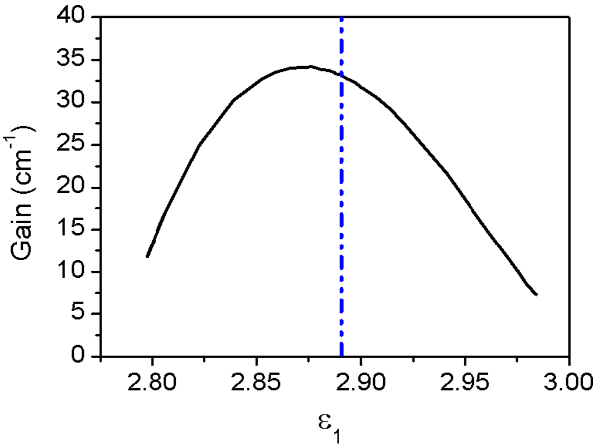

Figure 13. The solid line indicates the gain from the asymmetric sample structure as a function of permittivity, ε_1. The vertical line indicates the gain from the symmetric structure ($\varepsilon_1 = \varepsilon_3 = 2.89$) for comparison.

than the small increase in gain merits. Further, the overlap of this mode with the gain medium is likely to be poorer than for the standard LRSPP and this may well offset any reduction in loss when trying to achieve lasing threshold.

5. Conclusion

From an experimental point of view, the wide range over which emissive dipoles can be positioned and still produce gain means that a relatively thick dye layer can be deposited above a silver film. Preferably there would be a thin spacer layer between the two to prevent quenching of dye close to the silver. Provided all other sources of loss within the system are small, lasing may indeed be possible.

We have shown experimentally that emitters close to a metal film will lose power to both LRSPPs and SRSPPs. Using simulations, the power dissipated from an emissive dipole to both of these modes has been calculated as a function of metal thickness. It has been shown that despite loss of gain to the SRSPP, lasing from the LRSPP might still be possible.

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