

# Spontaneous emission from within a metal-clad cavity mediated by coupled surface plasmon–polaritons

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## Abstract

The emission of light from within a sub-wavelength metal-clad cavity is investigated. We find that emitters placed inside such a cavity emit into the coupled surface plasmon–polariton mode supported by this structure. We use grating structures to couple the non-radiative coupled surface plasmon–polariton modes to light and present the results of measurements of photoluminescence from such structures in the form of dispersion curves. We discuss our findings in the context of sub-wavelength cavities as a means to control the emission of light.

## 1. Introduction

The emission of light by excited species in the form of spontaneous emission depends not just on the species involved, but also on the local optical environment. More specifically the local optical environment must support a mode at the emission frequency [1]. If the density of such modes is high then the rate (probability) of spontaneous emission may be enhanced over that in free space, if the density is low then the emission may be inhibited [2, 3]; this is an area known as cavity quantum electrodynamics (CQED). Not only can the local optical environment be tailored so as to modify the rate of emission, the mode(s) into which emission takes place can also be used to direct emission into a laser mode and, if the structure is appropriate, act to direct the emission into the far field. This is an active area of research both for synthetic structures [4], and also for structures found in nature [5].

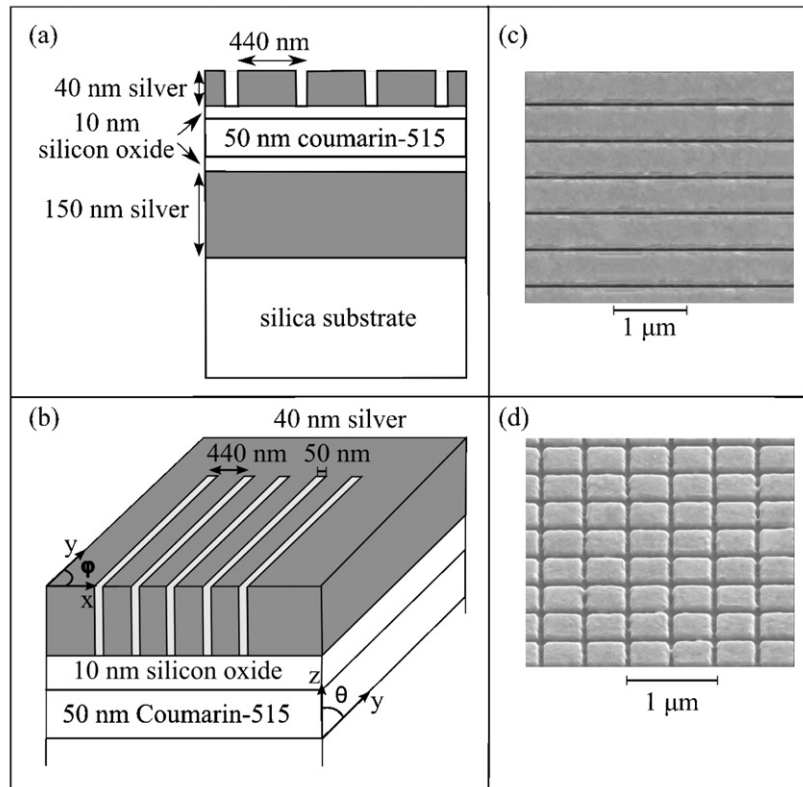
One of the attractions of using cavity QED is that one may gain greater control over the emitted light, as follows. Spontaneous emission is generally electric dipole in nature and as such there is only very moderate control possible over the direction of the emitted light—one requires control over the orientation of the dipole moment responsible for the emission. However, if the emission takes place into a mode of the local structure and the mode has a well defined wavevector then a number of techniques may be used to couple these modes to freely propagating light—the mode mediates the

emission. The momentum matching conditions that need to be satisfied for coupling such modes to light offer a way to control the momentum and hence direction of the emitted light, for example through the use of a grating.

One particularly effective set of modes that may be used to control spontaneous emission in this way are surface plasmon–polaritons [3, 6]. Surface plasmon–polaritons (SPPs) are optical modes that are guided by the interface between a metal and a dielectric. They have fields that decay away from the surface exponentially, and they result from the resonant interaction between an electromagnetic wave and the surface charges in the metal [7–10]. The near-field character of the fields associated with surface plasmons makes them well suited to interact with the near field of emitting species [3, 6]. Despite the relatively short propagation lengths of surface plasmon–polariton, periodic grating structures may be used very effectively to couple SPPs to freely propagating light [11–13].

Surface plasmon–polaritons exist not just on single metal–dielectric interfaces, but also in metal-clad cavities. Here the SPP modes associated with the two surfaces couple together (interact) to form two coupled SPP modes. These modes have attracted much attention recently because of the way in which their dispersion can be manipulated through control of the metals used and the cavity thickness [14–16]. It is not just the effective negative index associated with these modes that is of interest, the low frequency coupled SPP has no lower frequency cut-off. In the microwave regime this has

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**Figure 1.** Schematic diagrams (a) and (b) and scanning electron microscope (SEM) images of the sample structures; (c) shows an SEM of the single grating structure, (d) that of a bi-grating structure.

enabled  $\lambda/100$  cavities to be exploited as resonant absorbers of microwave radiation [17]. This approach has been extended into the visible region [18]. Also of potential importance is the fact that such sub-wavelength cavities can very significantly enhance the rate of spontaneous emission, by up to a factor of 25 [19]. We note that the lack of cut-off associated with cavity SPP modes means that they are also supported by metal–insulator–metal tunnel junctions [20] where the cavity thickness is of order 1 nm. The wavevector of cavity SPP modes in such structures is very high, and they have not successfully been exploited, though it is important in exploring tip enhanced spectroscopies from metal surfaces [21].

Despite their interesting attributes, the emission from species located within a sub-wavelength cavity and mediated by the coupled SPP mode supported by that cavity has received only limited attention. Here we report the results of a study with the aim of pursuing this investigation further, in particular we look at using grating structures to probe the way emission is mediated by the coupled SPP mode.

In what follows we first outline the concept behind our investigation. We then provide details of the sample fabrication. Measurements of spontaneous emission from our structures are then presented and discussed, finally, some conclusions are offered.

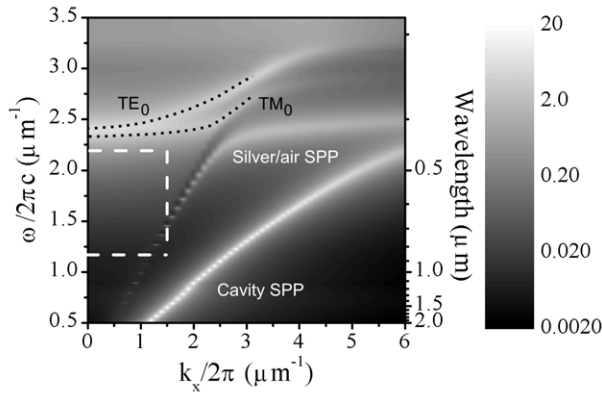
## 2. Conceptual overview and sample fabrication

Our purpose was to probe the way in which emissive species couple to the SPP modes of a thin (sub-wavelength) metal

cavity. Further, we wanted to test whether any modes other than SPPs were involved, and we wanted to look at how emission that takes place into the modes of the structure may be coupled to light. A schematic diagram of our metal-clad cavity structure is shown in figure 1.

We calculated the dispersion curve expected for planar versions of our structures, using the permittivity and layer thickness parameters given in figure 1, the dispersion curve is shown in figure 2. Light (white) regions in the figure represent strong emission by the source, thus allowing the modes to easily be seen and identified. The dispersion diagram was calculated by numerically evaluating the power dissipated by a dipole source located centrally in the cavity as a function of emission frequency and in-plane wavevector (momentum), the plane being that of the cavity. Details of such calculations are presented elsewhere [3, 22, 23]. The permittivity of the silver was taken as a polynomial fit to averaged literature data [24], whilst the dye layer and the silicon oxide layer were assumed to have a purely real permittivity of 2.89, based on other measurements of cavity reflectance and transmittance (not shown). The data in figure 2 indicate that emitters will couple to the coupled SPP mode roughly  $10\times$  more effectively than to the other cavity modes.

On the dispersion curve, figure 2, we have indicated with a dotted line the region of frequency/wavevector space that we were able to probe experimentally by recording photoluminescence from the samples as a function of emission angle and emission wavelength [25]. The dispersion curve shows four different modes. There is the coupled surface



**Figure 2.** Calculated power dissipation spectrum from a dipole source located in the centre of a planar version of the cavity structure shown in figure 1(a). White regions indicate significant power loss. The figure is plotted as a greyscale (arbitrary units), and as a function of frequency and in-plane wavevector. The different modes on this dispersion diagram are indicated. The area enclosed by a white dashed line indicates that part of parameter space that we were able to measure. The cavity was 70 nm thick and the top silver layer was 40 nm thick.

plasmon–polariton mode, a surface plasmon–polariton mode associated with the top silver–air interface, and two ‘standard’ cavity modes, one each of TE and TM polarization (labelled  $TM_0$  and  $TE_0$  respectively). Within our experimental window we can see that there are no modes that could act to mediate the emission from our structure, rather we will expect weak emission associated with luminescence that takes place directly through the top metal film.

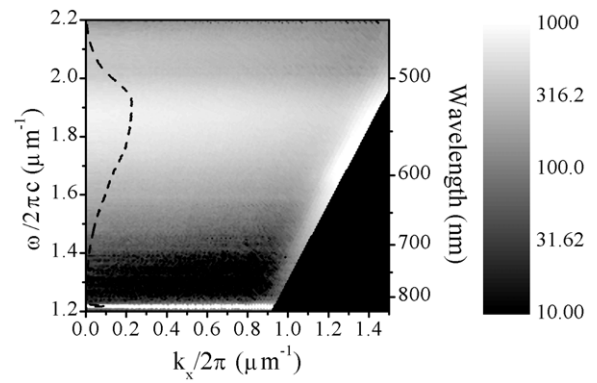
By adding a grating structure to the samples the modes outside the light cone may be coupled to freely propagating light, provided the grating pitch is chosen appropriately. The condition is that the wavevector of the mode,  $\underline{k}_{\text{mode}}$ , when scattered by an integer multiple of the grating wavevector,  $\underline{k}_g$ , where  $\underline{k}_g = 2\pi/\lambda_g$ ,  $\lambda_g$  being the pitch of the grating, satisfies,

$$|\underline{k}_{\text{mode}} \pm n\underline{k}_g| < k_0. \quad (1)$$

We first added a single grating to the structure with the aim of identifying the modes by which the emission of light is mediated. We then added a second grating to see what effect this would have on the emission characteristics.

The samples comprised an emissive layer of dye molecules on one side of which was an optically thick (150 nm) silver layer and on the other side a 40 nm silver layer. To protect the dye layer from subsequent processing it was bounded by 10 nm of silicon oxide on each side. All of these layers were deposited by evaporation under vacuum except the dye layer, this was instead deposited by spin casting from a chloroform solution. The upper silver layer was chosen so as not to be optically thick—this allowed us to probe emission from the cavity via light transmitted directly through this metal film. The thickness of the cavities produced was approximately 70 nm, significantly sub-wavelength (the emission of the dye, Coumarin 515, peaks at approximately 515 nm).

Later measurements required a grating to be added to the structure and this was accomplished by focused ion-beam



**Figure 3.** Photoluminescence (unpolarized, arbitrary units) data from the planar structure plotted as a function of frequency and in-plane wavevector. The feature at 820 nm is the second order of the pump beam. For comparison the dashed curve shows the emission spectrum of a bare dye film.

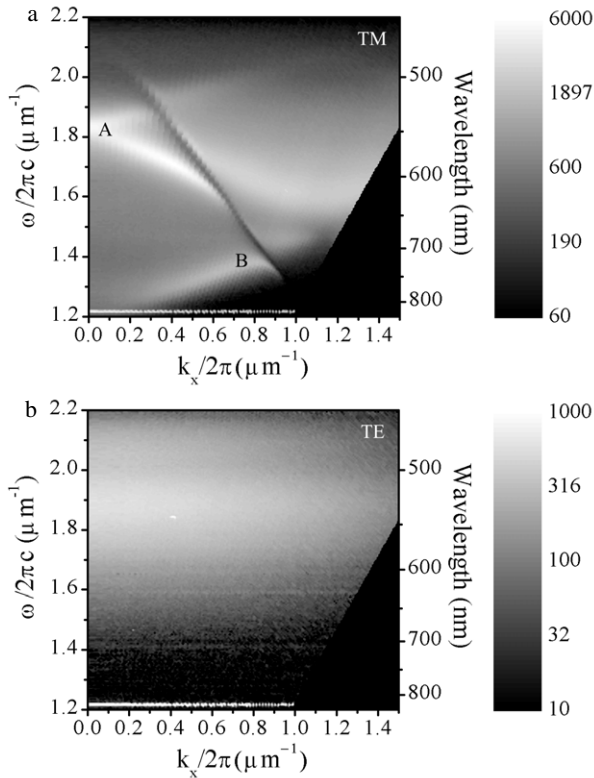
milling using an FEI Nova 600 system. Both single and bi-grating structures were produced, with the slots extending fully through the silver and having an approximately rectangular profile, see figures 1(c) and (d). Grating pitches were of order 500 nm and the area of the gratings was approximately  $200 \mu\text{m} \times 300 \mu\text{m}$ , sufficient to allow photoluminescence measurements to be acquired.

Photoluminescence (PL) measurements were performed by exciting the sample through the thinner silver film with a semiconductor laser operating at 410 nm. The resulting PL was collected with a lens (acceptance angle  $1^\circ$ ) and directed through an optical fibre to a CCD/spectrometer combination. Wavelength spectra were recorded as a function of the polar emission angle,  $\theta$  (as defined in figure 1(b)), in steps of  $1^\circ$  over an angle range of  $0^\circ$ – $50^\circ$ . Data were recorded both without and with a polarizer, thus enabling TE and TM polarized modes to be identified. The experimentally acquired data, PL intensity as a function of wavelength and angle ( $\lambda$ ,  $\Theta$ ) were converted to angular frequency and in-plane wavevector ( $\omega$ ,  $k$ ) thus allowing conventional dispersion diagrams to be presented.

### 3. Results and discussion

The PL (unpolarized) data from the planar cavity structure are shown in figure 3. As expected, we do not see any evidence for emission mediated by any of the modes shown in figure 2, rather the emission takes place directly through the top silver layer. The emission is relatively weak, and simply follows the free space emission spectrum of the dye (indicated in figure 3 for comparison).

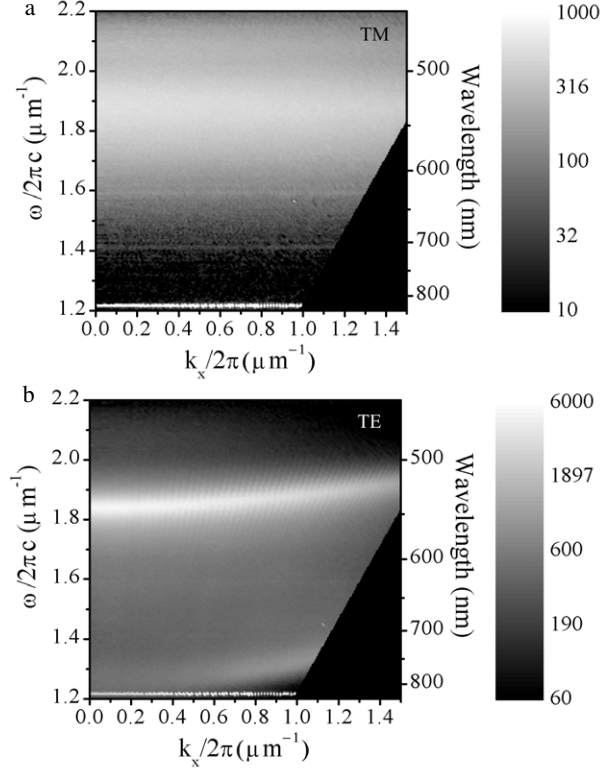
Data taken from the sample with a single grating structure imposed in the upper silver film (pitch 440 nm) is shown in figure 4. The grating was oriented so that the grating vector was in the emission plane (the grooves were perpendicular to the emission plane and parallel to the rotation axis), known as the classical mount. The upper set is for TM polarization, the lower for TE polarization. Looking first at data for the TE polarization, we see that it is almost identical to the data



**Figure 4.** The TM polarized ((a) upper) and TE polarized ((b) lower) PL (arbitrary units) collected from the sample with a single grating of pitch 440 nm at  $\phi = 0^\circ$ . Intensity is shown as a function of frequency and in-plane wavevector, white regions indicate strong photoluminescence.

obtained from the planar structure. This is to be expected—there are no TE polarized modes within the experimental window indicated on figure 2. In contrast the data for TM polarized emission shows strong features, indeed, the modal features dominate the emission—they are much stronger than the intrinsic emission of the dye. By considering how the modes indicated in figure 2 will be affected by scattering from the grating,  $\pm k_g$ ,  $\pm 2k_g$ , etc, by we can identify the modes indicated as A and B in figure 4 as the cavity SPP. Feature A corresponds to the cavity SPP scattered by  $+2k_g$ , whilst feature B corresponds to the cavity SPP scattered by  $-k_g$ . Note that the cavity SPP mode interacts with the scattered light-line when the two cross. Notice too that the cavity SPP mediated emission extends well outside the expected spectral range of the intrinsic emission spectrum, figure 3. This clearly shows how dramatically the emission mediated by modes alters the emission spectrum—it is much more than just a spectral filter for the emission.

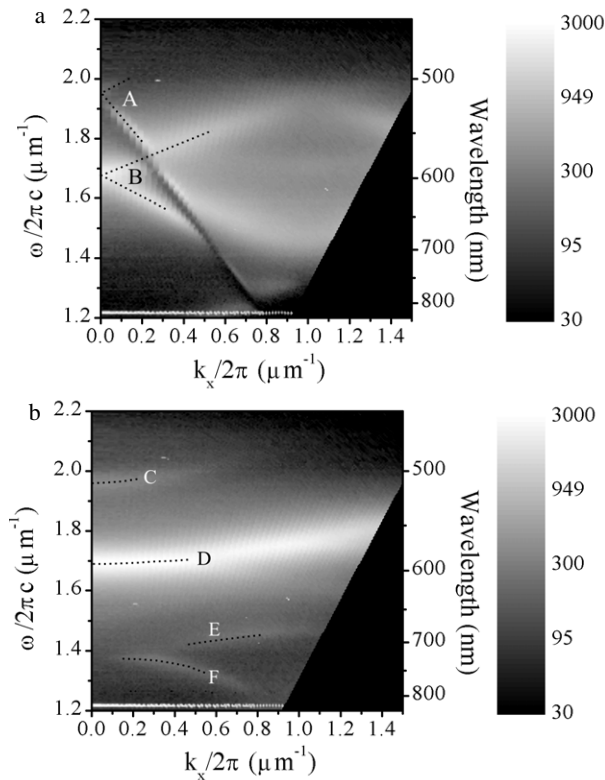
Somewhat surprisingly there is no evidence of any stop-band where counter propagating cavity SPP modes cross on the dispersion diagram. We had expected that such stop-bands might have occurred, they were seen before in a study on a similar (though not identical) structure [18]. The reasons for the lack of any stop-band are not clear, a fuller investigation would be required to explore this, possibly by varying the mark to space ratio of the etched grooves. However, such an exploration was beyond the scope of the present study.



**Figure 5.** The TM polarized ((a) upper) and TE polarized ((b) lower) PL (arbitrary units) collected from the sample with a single grating of pitch 440 nm at  $\phi = 90^\circ$ . Intensity is shown as a function of frequency and in-plane wavevector, white regions indicate strong photoluminescence.

The data shown in figure 4 were taken with the grating in the classical mount, i.e. the grating grooves parallel to the axis of rotation, the azimuthal  $\phi$  (shown in figure 1(b)) angle is  $0^\circ$ . If we rotate the sample by  $90^\circ$  so that  $\phi = 90^\circ$  we obtain the data shown in figure 5. The in-plane wavevector is now in an orthogonal direction to that associated with figure 4 so that the grating vector and the wavevector associated with the cavity SPP mode are now at  $90^\circ$  to each other. A direct consequence of this is that the cavity SPP mode is now seen in TE polarized PL emission, and shows only weak dispersion [26], whilst the TM polarized emission now shows no sign of the cavity SPP mode.

To see cavity SPP mediated emission for any polarization, and for all azimuthal angles, one can use two orthogonal gratings. To look at the effect of such a bi-grating structure we made another sample with grooves etched in orthogonal directions, shown in figure 1(d). The TM and TE polarized emission from this structure are shown in figure 6. The PL in both polarizations is now dominated by emission mediated by SPP modes. We can again identify the modes responsible for the different features by considering the dispersion diagram shown in figure 2 and how it will be affected by different scattering events. Emission mediated by the cavity SPP mode and scattered by various combinations of grating vectors appears as features B, D, E and F, whilst emission mediated by the silver–air SPP is seen as features A and C. Note that the modes seen in figure 6 for the crossed grating occur at slightly



**Figure 6.** The TM polarized ((a) upper) and TE polarized ((b) lower) PL (arbitrary units) collected from the sample with a bi-grating of pitch 500 nm. Intensity is shown as a function of frequency and in-plane wavevector, white regions indicate strong photoluminescence. The modes indicated are discussed in the text.

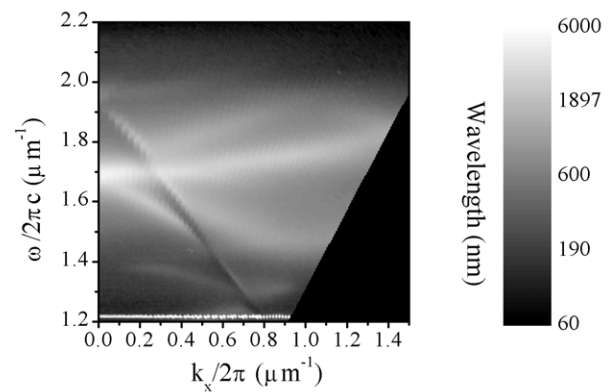
lower frequencies than the equivalent modes in figures 4 and 5 owing to the slightly longer pitch of the bi-grating, 500 nm cf 440 nm.

Finally, figure 7 shows the unpolarized emission from the bi-grating structure. There is strong photoluminescence, dominated by cavity SPP mediated emission. The modes involved can be inferred by considering a superposition of figures 6(a) and (b).

#### 4. Summary

Emitters placed in sub-wavelength metal-clad cavities such as those considered here couple most strongly to the cavity SPP mode, this mode is the one seen most strongly in figure 2. However, if the cavity is planar then little emission escapes by direct transmission through the metal film, emission mediated by the cavity SPP is not seen because this mode is non-radiative. The addition of a grating allows the momentum mismatch between this cavity mode and freely propagating light to be overcome, thus allowing cavity SPP mediated emission to be seen. The data presented here show that such emission is much stronger than directly emitted light.

For a single grating SPP mediated emission can not be achieved in all emission planes, but we have shown that this restriction can be overcome by adding a second grating that is orthogonal to the first. In conjunction with the much enhanced



**Figure 7.** Unpolarized PL (arbitrary units) collected from the sample with a bi-grating of pitch 500 nm. Intensity is shown as a function of frequency and in-plane wavevector, white regions indicate strong photoluminescence.

spontaneous emission rate associated with sub-wavelength metal cavities [19] there is clearly scope for using such cavities to greatly modify the spontaneous emission process using relatively easy to implement fabrication techniques. Such an approach may find application in light-emitting devices.

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