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Modification of the spontaneous emission rate of Eu^{3+} ions close to a thin metal mirror

R. M. Amos and W. L. Barnes
Thin Film Photonics Group, University of Exeter, Stocker Road, Exeter, United Kingdom

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We have experimentally determined how the spontaneous emission rate of Eu^{3+} ions depends upon both the distance from, and thickness of, a silver film. We show that as the silver film thickness is reduced (<100 nm) the well established influence of the mirror on the spontaneous emission rate is further modified. By comparing our data to theory we show that this is due to the availability of an additional decay channel, the surface-plasmon polariton on the far side of the metal film. [S0163-1829(97)03411-5]

I. INTRODUCTION

It is well known that the spontaneous emission rate is not a fixed property of the emitter, but that it depends on the local photonic mode density, as given by Fermi's Golden rule;[1,2] a fact first pointed out by Purcell.[3] In practice, the photonic mode density is modified by changing the boundary conditions of the electromagnetic field in the vicinity of the emitter. This concept is now well established and has lead to the field of cavity quantum electrodynamics.[4] The simplest alteration to the free space boundary conditions is a single reflecting surface. This was the system used by Drexhage *et al.*[5–7] in the first experimental verification of the modification of the spontaneous emission rate. They examined the lifetime of excited Eu^{3+} ions in front of a metallic surface, the distance between the ion and the mirror being varied in a precise manner to reveal the effect of the mirror on the spontaneous emission.

There are many ways in which the modification of the spontaneous emission may be viewed. We adopt here a well-tested classical approach as the basis of our theoretical calculations.[8] The emitter is considered to be a dipole oscillator, responding to its own field reflected from the mirror. Retardation of the reflected field plays a crucial role. If the reflected field returns to the emitter in the phase then the decay rate will be enhanced, if it is out of phase the decay rate will be suppressed. The distance of the emitter from the mirror is thus an important parameter in determining the spontaneous emission rate; the emission rate oscillates with increasing distance as the phase of the reflected field changes. An additional effect occurs when the ions are close ($\leqslant 1/4$ of the emission wavelength) to the mirror. The emission is quenched by resonant coupling between the dipole and the surface-plasmon polariton (SPP) mode that propagates at the surface of the metal mirror. At still closer distances the dipole can couple to lossy surface waves providing yet another quenching mechanism.[9,10]

The classical dipole approach allowed excellent agreement between experiment and theory to be obtained.[8] It further indicated that if the metal film was thin enough the excited ion could couple to the SPP on the far side of the metal film, resulting in a further increase of the excited-state decay rate.[8] The SPP modes of the two interfaces are distinct unless the dielectric constants of the media on either side of the metal film are similar. If this is the case and the metal film is thin enough, then the SPP modes associated with each interface may couple together to form a symmetric and antisymmetric coupled SPP pair.[11] The electric-field intensity of the symmetric mode is considerably enhanced over that of the single interface mode and, as has been calculated,[12] this can provide a significant further modification to the decay rate.

The effect of this second SPP decay channel has previously been observed by studying the transmission of molecular fluorescence through thin metal films.[13] However, decay rate studies have not been undertaken on such a system. We present here experimental results confirming the theoretical prediction[8] of the effect of the emitter coupling to the SPP mode of the far metal interface on the decay rate. We have done this by investigating the distance dependence of the emission lifetime of Eu^{3+} ions in front of silver mirrors, the latter having a thicknesses in the range 13 to 200 nm. In Sec. II we outline experimental details. Our results are presented and discussed in Sec. III, the associated theory being outlined in Sec. IV. The work is summarized in Sec. V where we also offer possible directions for future research.

II. EXPERIMENT

To examine the effect of a nearby thin metal mirror on the emission lifetime of a nearby emitter we adopted a procedure very similar to that used in the pioneering work of Drexhage *et al.*[5–7] Eu^{3+} ions were used as the emitters since they emit in the visible, ~ 614 nm, and have a conveniently long lifetime, ~ 1 ms. In addition, the transition giving rise to the fluorescence ($^5D_0 - {}^7D_1$) is primarily electric dipole in nature allowing for a simple theoretical treatment of the system. Silica flats, polished to $\lambda/10$, were cleaned and rendered hydrophobic by exposure to 1, 1, 1, 3, 3, 3-hexamethyldisilazane vapor for 12 h. A silver film (99.99% pure) was then deposited by thermal evaporation onto the surface to form the mirror. The thickness of the film was monitored by a quartz-crystal oscillator and the film deposited at a rate of 100 nm per minute under a background pressure of 2×10^{-6} Torr.

The Langmuir-Blodgett (LB) technique[14] was used to deposit a known number of optically transparent organic monolayers of 22-tricosenoic acid onto the silver film, these formed the spacer layer between the mirror and the emitters. The LB technique provides a precise way of controlling the emitter/mirror separation. A 0.2 mg ml^{-1} solution of 22-tricosenoic acid in chloroform was spread on a pure water subphase (pH 5.5 and resistivity 18 MΩ cm^{-1}). The resulting film was compressed to a surface pressure of 30 mN m^{-1} and deposited onto the silver surface with Y-type deposition at a dipping speed of 0.2 mm s^{-1}.

The emitters were added by dipping one further LB monolayer onto the sample, the film being a europium-doped chelate. The chelate serves two functions; first it provides a convenient way to incorporate the Eu^{3+} ion into an LB film and second, the organic ligands provide a large absorption in the UV from which the Eu^{3+} ion may be excited by nonradiative intramolecular energy transfer. This latter aspect is useful since rare-earth ions have, in general, rather small absorption cross sections. The chosen chelate, hexadecyl pyridinium tetrakis (1,3-diphenyl-1,3-propandione) europium, was such that the Eu^{3+} ion resided in a chemically symmetrical environment. This ensured that the orientation of each ion in the film was on average random, thus allowing easier interpretation of the data.

The europium chelate did not form a stable Langmuir film under compression, collapsing at surface pressures greater than 10 mN m^{-1}, due mainly to crystal growth in the film. High quality monolayers could not therefore be deposited with the usual vertical dipping techniques. Instead, the monolayer was transferred to the substrate horizontally at a pressure of 5 mN m^{-1}, thus avoiding film collapse. The film was spread from a solution containing 1 mg of the chelate in 1 ml of acetone and 4 ml of benzene. The emission intensity of the europium chelate suffers from photobleaching. The lifetime, however, is unaffected so that for the study reported here the photobleaching did not present a problem.

The thickness and dielectric constant of the silver films used as mirrors were measured by monitoring the angle-dependent reflectivity of p-polarized light at 632.8 nm.[15] The substrates were index matched (on the glass side) to a silica prism, the reflectivity being recorded for angles of incidence near the critical angle. By fitting theory, based on multilayer Fresnel reflection coefficients[16] to the experimental data the dielectric constant and thickness of the silver films were determined.

The excited-state lifetime of the Eu^{3+} ions was measured using standard photon counting techniques. The sample was excited by 5-ns pulses of UV light (337 nm) from a nitrogen laser. The resulting fluorescence was collected by a lens and focused onto the entrance silts of a spectrometer set to pass 614 nm. The emerging light was focused onto a photomultiplier (Hamamatsu R955), cooled to −30 °C. The overall time resolution of the system was ~5 ns. The output pulses from the photomultiplier were analyzed by a multichannel photon counter (Stanford SR430) and logged by a computer.

III. RESULTS

The time evolution of the fluorescence was well approximated by a single exponential, provided the fabrication procedures outlined above were followed. The emission lifetime was determined from the experimental data by fitting a function of the following form to the data.

$$I(t)=I_0\exp(-t/\tau)+I_b, \qquad (1)$$

where I_0 is the initial count rate of the recorded fluorescence signal, τ is the excited state lifetime of the Eu^{3+} ions, and I_b the background count rate. Generally the background level was less than 1 count and the error in the fitted lifetimes less than 1%. Two sample decay curves are shown in Fig. 1, together with the fitted theoretical curves. The quality of the exponential fits indicates excellent uniformity of the LB spacer layers.

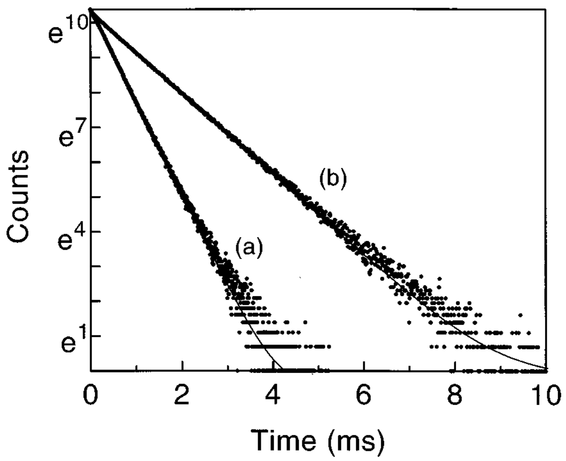

FIG. 1. The time evolution of fluorescence from Eu^{3+} ions situated at 31.2 nm from a (a) 13.3-nm-thick silver mirror, and (b) from a 200-nm-thick silver mirror. The corresponding exponential fits of the functional form expressed in Eq. (1) are also shown. The intensity of the emission for case (a) was an order of magnitude smaller than that of (b).

Optically thick (~200-nm) silver mirrors were coated with spacer layers of 22-tricosenoic acid using the LB technique. In order to decrease the number of samples needed each mirror had 16 different spacer layer thicknesses deposited onto it. Thus by multiplier dipping only four samples were required to obtain the data shown in Fig. 2. The lifetime was determined for mirror-emitter separations from 5–500 nm.

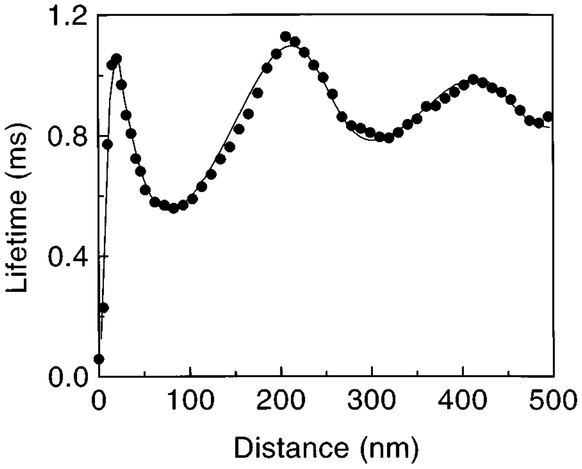

FIG. 2. The excited-state lifetime of Eu^{3+} ions as a function of the distance from a 200-nm-thick silver mirror. The solid line is the theoretical dependence based on the classical dipole model outlined in the text.

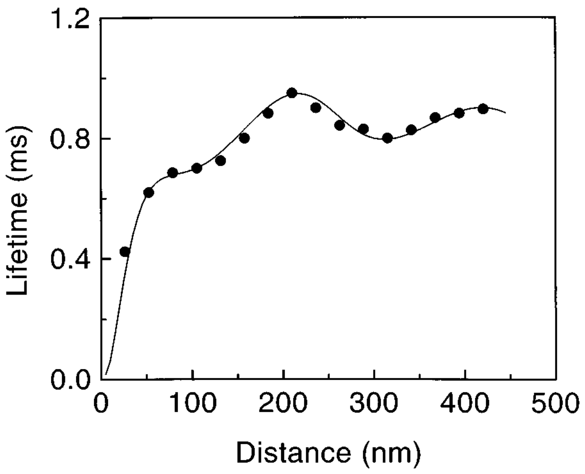

FIG. 3. The excited-state lifetime of Eu^{3+} ions as a function of the distance from a 14-nm-thick silver mirror. The solid line is the theoretical dependence based on the classical dipole model outlined in the text.

The results of these measurements are shown in Fig. 2. They are in excellent agreement with those of Drexhage[7] and provide a good validation of our experimental technique. The spontaneous emission lifetime modifications produced by the mirror as discussed in the Introduction are clearly seen. For distances greater than ~30 nm the lifetime oscillates as the phase of the reflected field changes, the modulation decreasing as the emitter/mirror separation increases, as expected by considering the asymptotic limit. For small separations, <30 nm, the lifetime is rapidly quenched, primarily by coupling to the SPP mode at the silver/LB interface. The solid curve in Fig. 2 shows the calculated distance dependence of the lifetime, based on the classical model discussed in the following section. For the moment we just note the good agreement between experiment and theory.

We next examined the effect of reducing the thickness of the silver mirror. Figure 3 shows the experimentally determined distance dependence of the emission lifetime above a 14-nm-thick silver film, significantly different from the results obtained for the optically thick silver film of Fig. 2. Before looking at further experimental results it is worth considering the three main effects that take place as the silver thickness is reduced. First, the reflectivity of thin films is lower than that of thick ones, resulting in a reduction in the amplitude of the reflected field at the emitter site. This in turn results in a reduction in the amplitude of the distance dependent lifetime oscillations. Second, as the silver thickness is further reduced the dielectric response of the silver eventually changes, due to the way in which the silver is deposited during the thermal evaporation process. For very thin films the silver growth on the silica substrate results in an islandized film.[17] The effective dielectric constant is thus different from the value for thick silver films. The real part is somewhat reduced and the imaginary part greatly increased, see Table I. This change in the dielectric constant greatly effects the nature of the associated SPP mode. In particular, the increased imaginary component of the dielectric constant results in a broadening of the SPP resonance, giving rise to a net increase in the coupling between the SPP and the emitting dipole. Quenching due to the SPP mode is therefore increased. Third, when the silver is very thin, a significant fraction of the dipole field can penetrate the silver and couple to SPP modes at the silver/silica interface. This again provides further quenching when the silver film is thin.

TABLE I. The silver dielectric constant and thickness of the different mirrors used. The dielectric constants at 632.8 nm (measured) and at 614 nm (inferred) are given. The values for the 200-nm-thick film agree well with those found in the literature (Refs. 25, 26, 27, and 28).

Thickness of silver (nm) ±0.5 nm	Measured ε_r at 632.8 nm ±0.1	Measured ε_i at 632.8 nm ±0.05	ε_r used in model	ε_i used in model
200.0	−17.5	0.65	−16.0	0.4
66.7	−17.1	0.63	−15.5	0.4
46.1	−17.2	0.60	−16.0	0.4
38.4	−15.9	0.94	−14.2	0.7
26.7	−16.5	0.97	−15.0	0.7
13.3	−9.1	16.0±1.0	−9.1	16.0

The experimental results in Fig. 4 demonstrate how the excited-state lifetime of the Eu^{3+} ions depends upon the distance from silver films of different thicknesses. Each silver film was characterized as before by recording angle-dependent reflectivity and fitting theory to it. We clearly see in Fig. 4 that reducing the silver thickness has a marked effect on the recorded lifetimes. To understand the physics behind these changes it is instructive to model the lifetime data theoretically.

IV. THEORY AND FURTHER DISCUSSION

Considerable theoretical work has been devoted to studying how the spontaneous emission rate of an excited atom is modified by the presence of a reflecting surface. A convenient and successful approach is to treat the atom as a classical dipole oscillator,[8,9,18–21] much as was done originally

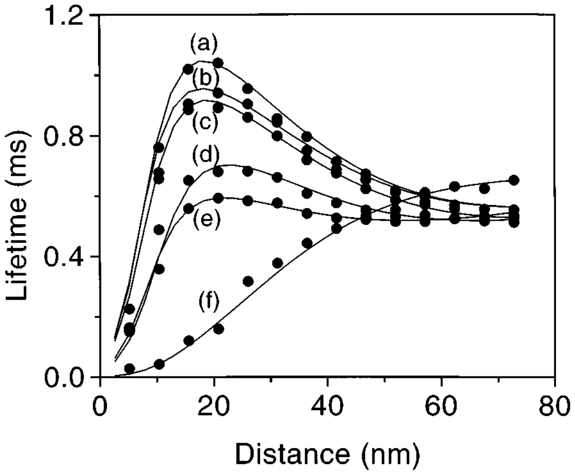

FIG. 4. The excited-state lifetime of Eu^{3+} ions as a function of both the distance from, and the thickness of, a silver mirror. The corresponding silver thicknesses are (a) 200 nm, (b) 66.7 nm, (c) 46.1 nm, (d) 38.4 nm, (e) 26.7 nm, (f) 13.3 nm, respectively.

by Sommerfeld in studying the effect of the earth on radio wave propagation.[22] The problem reduces to finding the reflected field at the position of the dipole due to the presence of the various interfaces, with the retardation being taken into account. This may be done by expressing the dipole field as an expansion of cylindrical[8] or plane[18,19,21] waves. The modified decay rate can then be written as[8]

$$b_{\perp,\parallel}=b_0(1-qz_{\perp,\parallel}), \tag{2}$$

where b_0 is the radiative decay rate in the absence of any interfaces and q is the radiative quantum efficiency. The parameters $z_\perp$ and $z_\parallel$ are given by

$$z_\perp=1-\left(\frac{3}{2}\right)\mathrm{Im}\int_0^\infty\frac{(1-r_{12}^p e^{-2\beta_{12}})(1-r_{13}^p e^{-2\beta_{13}})}{(1-r_{12}^p r_{13}^p e^{-2(\beta_{12}+\beta_{13})})}\frac{u^3}{l_1}du, \tag{3}$$

$$z_\parallel=1-\frac{3}{4}\mathrm{Im}\int_0^\infty\left\{\frac{(1+r_{12}^s e^{-2\beta_{12}})(1+r_{13}^s e^{-2\beta_{13}})}{(1-r_{12}^s r_{13}^s e^{-2(\beta_{12}+\beta_{13})})}+(1-u^2)\frac{(1+r_{12}^p e^{-2\beta_{12}})(1+r_{13}^p e^{-2\beta_{13}})}{(1-r_{12}^p r_{13}^p e^{-2(\beta_{12}+\beta_{13})})}\right\}\frac{u}{l_1}du, \tag{4}$$

where $r_{12}^{p,s}$ and $r_{13}^{p,s}$ are the Fresnel reflection coefficients for p and s polarized light at the nearest interface below and above the dipole, respectively; $r_{12}^{p,s}$ takes into account the reflection arising from the silver/silica interface. The exponents β_{12} and β_{13} are the phase changes due to the distance between the nearest interface below and above the dipole, respectively. The parameter u is the component of the wave vector in the plane of the interface, k_x, normalized with respect to magnitude of the far-field wave vector of the dipole radiation field, and $l_1=-i(1-u^2)^{1/2}$. The integration takes place over all positive values of u.

We have assumed the dipole orientation to be random (isotropic), a reasonable assumption since we chose the chelate specifically so that the dipole moment of the Eu^{3+} ion would have no preferred orientation, we can thus write the decay rate for an isotropic distribution of dipole orientations, b_{iso},

$$b_{\mathrm{iso}}=\frac{2}{3}b_\parallel+\frac{1}{3}b_\perp\,. \tag{5}$$

We also assume that the LB layers are both nonabsorbing and isotropic. Both these assumptions are known to be untrue[23] but, as we shall see below, they provide a good enough approximation for the present study. The thickness of the 22-tricosenoic acid monolayers was determined to be 2.6 nm using the critical edge technique[15] in good agreement with previous measurements.[23] The chelate layer was assumed to have the same thickness as a monolayer of 22-tricosenoic acid, a reasonable assumption given the structure of the two molecules. We further assumed that the Eu^{3+} ion was in the middle of the chelate layer, thus placing it 1.3 nm below the top surface of the sample. Although this is a rather short distance it is nonetheless important in obtaining a good match between experiment and theory.

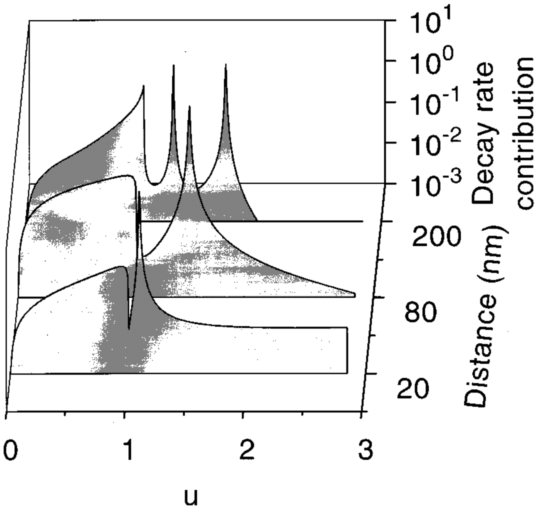

FIG. 5. The contribution to the spontaneous emission rate [given by Eq. (5) in the text] as a function of u for a 200-nm-thick silver film. Data for dipole-mirror distances of 20, 80, and 200 nm, are shown, respectively.

The integrands of Eq. (5) [specified in Eqs. (3) and (4)] give the contribution to the spontaneous emission rate at a particular value of u. For $0\leqslant u\leqslant 1$, the contribution represents emission of radiation into the half space above the sample. The contribution from $u>1$ corresponds to evanescent, high wave-vector components of the dipole field. They do not, therefore, propagate into the far field unless they can couple to guided modes of the system. If the LB film is thick enough it may support waveguide modes. It is important to take into account the power lost to these modes in order to obtain the correct lifetime dependence. The coupling to these modes, however, occurs at the expense of the free radiation and provided that the LB film is nonabsorbing the lifetime is unaffected.[24]

Coupling may also occur between the dipole field and the SPP at the silver/LB interface. In the absence of loss in the mirror this would not make any contribution to the spontaneous emission rate. However, for a real, lossy mirror, resonant coupling to the SPP mode is a power dissipating mechanism resulting in an increase in the spontaneous emission rate. Because the fields, associated with the SPP mode and the high wave-vector components of the dipole field are evanescent, significant coupling between the two only takes place at relatively small separations, below ~50 nm.

Figure 5 shows the contribution to the spontaneous emission rate given by the integrand of Eq. (5), as a function of u, for the 200-nm-thick silver mirror system. Three mirror/dipole separations are shown, 20, 80, and 200 nm. At a distance of 200 nm the excited state lifetime is dominated by coupling to photons, both free and guided. Coupling to the SPP mode at $u\approx 1.7$ is very small since the overlap of the SPP and appropriate dipole field is very weak at this separation. The contribution at $u\approx 1.3$ is due to coupling between the dipole field and the lowest-order TE waveguide mode of the silver/LB/air system. At a separation of 80 nm the coupling to the SPP is much greater and the SPP mode has shifted to lower momentum, $u\approx 1.43$, due to a reduction in the effective dielectric constant of the LB film experienced

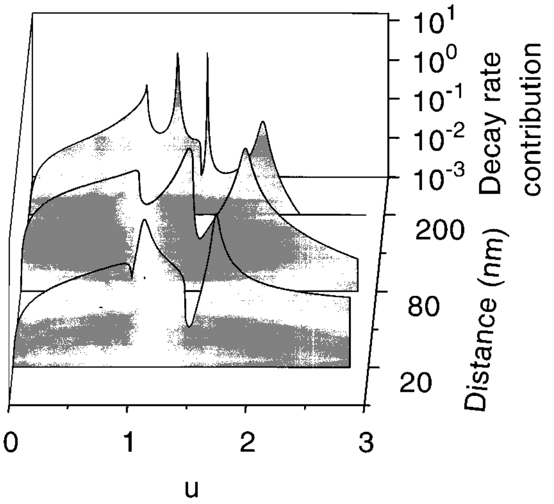

FIG. 6. The contribution to the spontaneous emission rate [given by Eq. (5) in the text] as a function of u for a 26.7-nm-thick silver film. Data for dipole-mirror distances of 20, 80, and 200 nm, are shown, respectively.

by the SPP mode. This spacer layer thickness is such that there are no waveguide modes present. At a separation of 20 nm coupling to SPP modes dominates the decay rate.

Figure 6 shows a similar set of calculated data for a thin silver mirror 26.7 nm thick. Again, at a separation of 200 nm the spontaneous emission rate is dominated by the contribution due to the emission of free and guided radiation. The coupling to the SPP at the silver/LB interface, $u \approx 1.55$, is small. Notice also the very small feature at $u \approx 1.95$, it represents coupling of the dipole field to the SPP mode at the lower surface of the metal, i.e., the silver/silica interface, the coupling is very weak at this separation. On reducing the separation to 80 nm the coupling to both SPP modes is considerably increased due to the greater overlap between the dipole and SPP fields. When compared to the results for the thick silver film, Fig. 5, the SPP resonances are now seen to be considerably broader. This is because of the higher value of the imaginary part of the dielectric constant of the silver for this thickness of silver film, 0.7 compared to 0.4 for the 200-nm film, Table I.

Performing the integrations in Eq. (5) enables the modified spontaneous emission rate, and hence the lifetime, to be calculated and a comparison made between the experimental data and theory, as shown in Figs. 2, 3, and 4. In fitting theory to data for the thick silver mirror, Fig. 2, we were able to determine the radiative quantum efficiency to be 0.7. The effective dielectric constant of the LB film was taken from critical edge measurements to be $\varepsilon_r = 2.49$, in good agreement with what might be expected from knowing the full biaxial form of the dielectric tensor of the material.[23] The adjustable parameters in the theory were thus reduced to the thickness and dielectric constant of the silver films only.

The theory traces in Fig. 4 made use of the silver parameters derived from fitting the reflectivity data, Table I. The dielectric constants for the lifetime modeling have been shifted slightly from the fitted values to take into account the dispersion between the characterization wavelength, 632.8 nm and the emission wavelength of the Eu^{3+} ions, 614 nm. These are also shown in Table I. The dielectric constant of the 22-tricosenoic acid was assumed to be dispersionless over this range.[23] The good agreement between experiment and theory would seem to justify the approximations we have made in undertaking our calculations, in particular, that of using an effective dielectric constant for the inhomogeneous thin silver films.

V. SUMMARY

We have shown experimentally how the spontaneous emission rate of a Eu^{3+} ion depends upon the distance from, and the thickness of a thin silver mirror. By comparing our data with a theoretical model we have been able to identify the mechanisms responsible for the observed changes. As the thickness of the silver film is reduced a new decay channel is opened up through coupling of the dipole field to the SPP mode on the far side of the mirror. Additional changes to the emission rate arise for very thin silver films due to the change in the dielectric constant of such films.

Many interesting possibilities are prompted by this work. Introducing a corrugation to the surface can allow enhanced coupling of the dipole field to the SPP modes of both interfaces.[29] In fact, by using an appropriate combination of corrugations it is possible to completely block the SPP mode on a particular surface[30] in much the same way as the propagation of light may be prohibited in a photonic band-gap material.[31] The possibility exists therefore of either blocking the quenching due to the SPP decay channel, or enhancing it by ensuring the emission frequency of the emitter lies at the edge of the SPP band gap.[32] A more extreme modification may be induced if the inhomogeneous nature of the very thin silver films is taken still further so that the film becomes islandized. In this case the asymptotic resonance frequency of the SPP mode, which is usually at ~3.6 eV, can be lowered. If this frequency could be lowered such that it becomes resonant with the emission frequency of the emitter then one would expect very significant quenching to take place. The effect of such islands on the frequency of emission, rather than the emission lifetime, has already been observed, albeit for a rather different system.[33] We are vigorously pursuing these interesting ideas.

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