

# Probing semiconductor confined excitons decay into surface plasmon polaritons

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**Abstract** The study of the interaction of surface plasmon polaritons (SPPs) with quantum emitters has become very important in the last few years. The ability to design optical devices as well as investigate the physics of strongly interacting systems is some of its useful applications. In this paper, we will show some results on the decay of excitons confined in a InAs/GaAs quantum dot into SPP modes confined in a metallic thin film made of Au, an important step toward the investigation of the basic features of the SPP–exciton interaction. The lifetime of the ground state level was investigated and shown to decrease with the presence of the metallic film.

## 1 Introduction

Plasmonics [1] has evolved in the last few years as a promising candidate to substitute electronics [2]. The main important features present are the possibility to design optical devices at the visible range [3–5], i.e., operating at frequencies around THz, as well as the high confinement that can be achieved by exploiting the unique features of the so-called surface plasmon polaritons [6].

The SPP waves are a result of the interaction between light and free electrons in a metal/dielectric interface, which results in the confinement of light that propagates in a two-dimensional space with evanescent electromagnetic

fields penetrating both in the metal and dielectric media. As a result of this confinement, the electromagnetic field (EMF) can be enhanced by many orders of magnitude. These high EMF strongly interact with the medium in which it propagates, and due to the losses present in the metal these waves display a finite propagation length [7].

In order to enhance the propagation length of SPPs, a common approach relies on the idea of doping the dielectric medium with a gain media having long-lived energy states [8], compared to SPP lifetime, as a route to suppress the losses in the interacting wavefunction of the SPP. In these situations, it can be achieved a strong coupling regime between SPP waves and the quantum emitters [9, 10] present in the gain media, an essential ingredient which may provide new exotic features of photons interactions such as the so-called quantum fluids of light [11].

A way to probe this strong interaction can be inferred by the modification in the lifetime of the quantum emitters which is affected by the interaction with the SPPs. When placed near metallic nanoparticles (NPs), excitons in a quantum dot (QD) or quantum well (QW) decay into light and also in localized SPPs (LSPPs) [12]. Under some restrictive conditions, it was observed that the photoluminescence (PL) spectra may increase or decrease depending on the density of these NPs [13, 14]. However, the relative position between the quantum emitters and the metallic NPs is not well controlled in these systems. In our case, we have focused on the control of the distance between the quantum emitter and the metal/dielectric interface.

The quantum emitters are a single layer of InAs quantum dots embedded in a GaAs matrix with variable cap layer. The precision on the cap layer thickness is on the order of an atomic layer which can be achieved with molecular beam epitaxy (MBE) technique. The metallic film is a thin layer of 6 nm made of gold. We performed a

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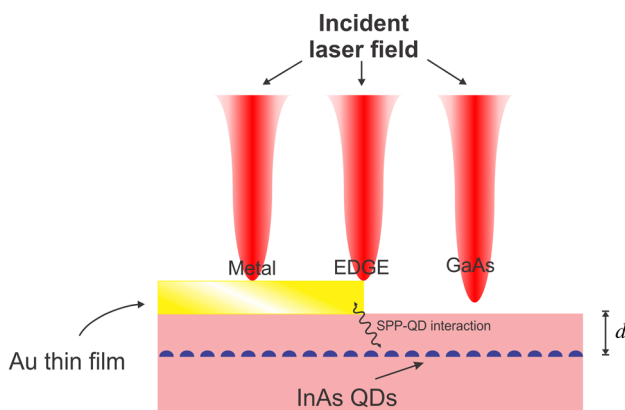
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time-resolved photoluminescence (TRPL) investigation to obtain the lifetime of the QDs as a function of the cap layer thickness in a region where the GaAs matrix is covered with the metallic thin film and compared to the lifetime when there was no metallic film over the sample. In Fig. 1, we present a scheme of the measurement where the interaction of SPPs with QD excitons is shown to influence only the QDs near the Au/GaAs interface. In the present paper, we show that the SPP–QD coupling mechanism is responsible for a strong decrease in the lifetime of exciton in the QD.

## 2 Method and results

The sample was grown by molecular beam epitaxy on (100)-oriented GaAs substrate. After the desorption of the GaAs oxide, a 300-nm buffer layer was grown at a substrate temperature of 580 °C. After the buffer, we deposited 2.0 monolayers (ML) of InAs at 530 °C followed by a 30 s annealing. Then a cap layer with thickness  $d$  of 10, 20, 30, 40 and 50 nm of GaAs was grown. The minimum value of cap layer is enough to guarantee that the InAs QDs are completely covered with GaAs although surface states play an important role in the decay mechanism for  $d = 10$  nm.

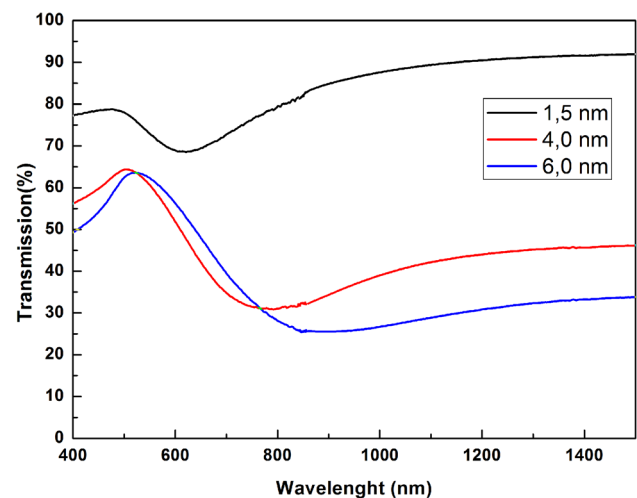
After the dielectric doping with QDs, we deposited a thin layer of Au over the GaAs surface using an evaporation chamber. A mask was used to produce an area without the film. The film was made thin enough in order that a photoluminescence (PL) signal can still be obtained when the laser beam is focused over the Au film. However, for a very small thickness the film consists of an agglomerate of Au NPs. In order to ensure that this is not the case for our sample, the deposition of the metallic film was made both



**Fig. 1** Scheme representing the photoluminescence of InAs QDs in a GaAs matrix covered and uncovered with a thin gold film of 6 nm width. The figure also represents the competition between the radiative decay and SPP decay for a quantum emitter placed near a metallic film

on the GaAs surface and on a glass substrate with index of refraction 1.5. We employed the glass in order that the transmission of light through the metallic film can be analyzed also for the visible range, which is not possible for GaAs due to its bandgap energy (around 1.42 eV) which corresponds to 870 nm.

The transmission was obtained for some values of thickness ( $h = 1.5, 4.0, 6.0$  nm), and the results are shown in Fig. 2. The absorption peak at 618 nm for  $h = 1.5$  nm is due to the absorption of light by the Au NPs [15], and the high value of transmission (around 90 %) above this peak indicates no formation of the Au thin film. For  $h = 4.0$  nm, the peak is shifted to 775 nm, as a result of the increment in the NPs size, and the value of transmission at the infrared range shows that the film was formed. However, as the peak on 775 nm is still sharp, the Au NPs may generate LSPPs and play an important role in the decay mechanism due to its high electromagnetic field in its vicinity. On the other side, a thicker film with  $h = 6.0$  nm does not presents this sharp absorption peak due to NPs, indicating the formation of an almost smooth film, and the transmission around the QD exciton energy (near 1100 nm) is large enough to ensure that enough PL intensity can be obtained, the reason why this thickness was used in the following experiments. It is worthwhile to mention that when above the GaAs substrate, which has a higher index of refraction, the absorption peak should be shifted to smaller wavelength when compared to the glass substrate; this ensures that no influence of the NPs on the QD energy states is expected.



**Fig. 2** Optical transmission of light for a sample of glass covered with a thin Au film with thickness  $h = 1.5, 4.0, 6.0$  nm. The peak around 500 nm is due to the bulk plasmon of Au. The absorption peak that shifts for infrared with a 6 nm Au film made over a glass substrate. The absorption around 800 nm corresponds to a LSPP mode in the agglomerate of NPs

The PL measurements were taken at 7 K with a closed-cycle cryostat. For excitation, we used a 730 nm diode laser guided by a multimode optical fiber with 50  $\mu\text{m}$  core to the sample, with a spot diameter of 70  $\mu\text{m}$ . The optical excitation power varied in the range of  $10^{-3}$ – $10^1$  mW. However, the most robust results were obtained for a laser power ranging from  $10^{-1}$  to  $10^1$  mW. The PL emitted light was collected by another optical fiber and guided to a 0.5 m focal length spectrometer entrance and detected by an InGaAs photodiode detector array. Figure 3a shows the normalized CW PL spectra of the InAs/GaAs QDs samples measured on the region without the Au layer (on GaAs) for different GaAs capping layer thickness. The average height of InAs QDs is approximately 8 nm with a pyramidal shape of 20 nm base. For GaAs capping layers larger than the 10 nm, the peak position is  $\sim 1.10$  eV with a linewidth in the range of 32–38 meV. The peak position of the sample with 10 nm capping layer is shifted to 1.08 eV and the with a linewidth of 45 meV. The QDs capped with 10 nm of

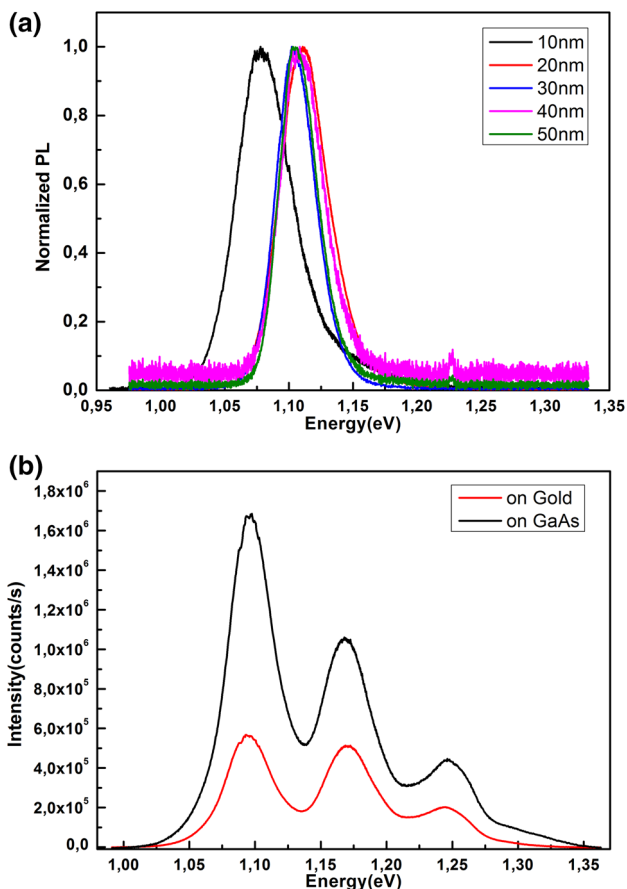
GaAs are fully capped [16], but the strain field that remains on the 10 nm layer shifts the QD ground state to lower energies. The large surface potential (around 5 eV) does not affect significantly the energy levels of the QDs. The reduction in the emission intensity for the 10 nm capped sample is due to the strong nonradiative recombination at the GaAs/air surface. The lifetime of carriers of the QDs is shortened also due to the nonradiative recombination at the surface. The charge transfer between the QDs and the surface states decreases significantly for GaAs capping layers larger than 20 nm.

For a comparison, Fig. 3b shows the PL spectrum for the sample with  $d = 50$  nm at both regions, with and without Au layer, at 5 mW (120 W/cm<sup>2</sup>) laser power. The peaks at 1.17 and 1.24 eV are the first and second QD excited states [17]. To ensure that the distribution of QDs is almost the same when the laser is on both regions, we observe that at the edge of the film, as shown in Fig. 1, the integrated signal decreases monotonically to a constant plateau. The first signature of interaction between the SPPs and the QDs is the change in the ratio of the signal from first excited to ground state QD energy when the laser is focused onto the Au layer. The presence of the Au film induces a more pronounced formation of the first excited state due to a higher coupling factor between the SPPs and the exciton than for the ground states. The reason for the higher value of the coupling factor is that mesoscopic effects of the exciton wavefunction play a major role for excited states [18].

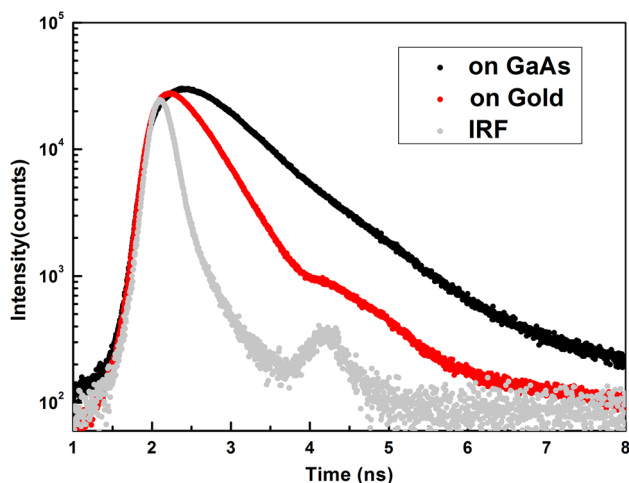
To show that this coupling influences not only the relaxation mechanisms of the carriers in the GaAs matrix, using the same setup as before, we also performed TRPL investigations by setting the 730 nm diode laser to pulsed mode with a 70 ps pulse width and a repetition frequency of 80 MHz. The lifetime measurements were performed following the peak position of ground state as function of excitation power. The internal response function (IRF) of the system was measured, with a resolution to better than 100 ps.

Figure 4 shows a single measurement of TRPL signal for sample with  $d = 30$  nm thick GaAs, at a laser power of 1 mW, measured on GaAs and on Au regions. In this plot, we show that the decay slope for measurements on GaAs is lower than that on gold, which means a higher lifetime of carrier recombination. The instrument response function of the system is also present, and its lower intensity peak around 4 ns coincides with the shoulder on the Au data. Fits of decay slopes were performed (not shown here) in order to acquire the lifetime.

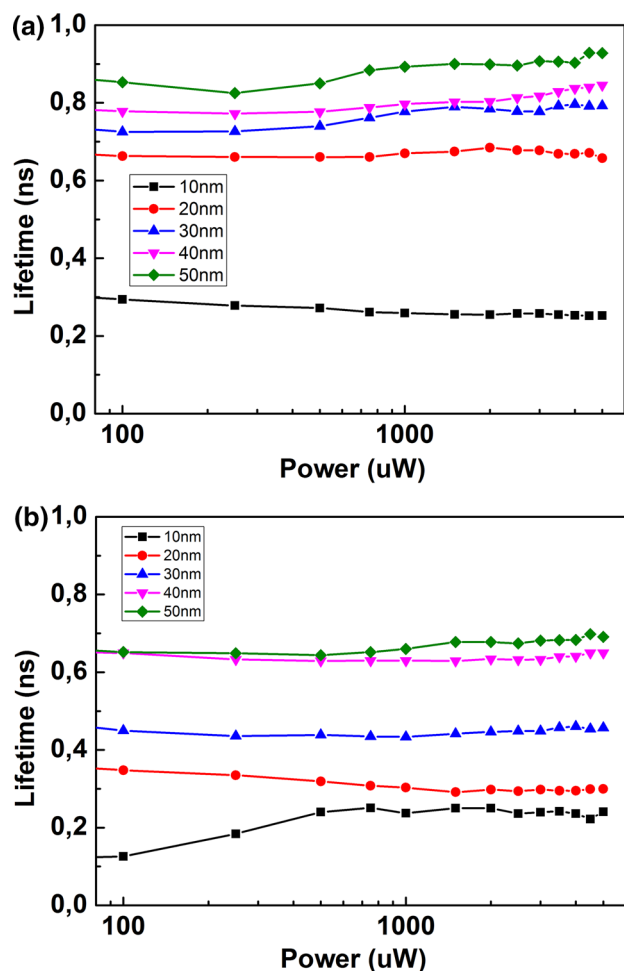
The results obtained for the lifetime of the ground state energy in the QD are shown in Fig. 5 for the region without (a) and with (b) the Au film. The data were taken for laser power varying from 100  $\mu\text{W}$  (0.2 W/cm<sup>2</sup>) to 5 mW



**Fig. 3** a Normalized PL signal for samples between  $d = 10$  nm to 50 nm of GaAs capping layer thickness without Au film taken at 10  $\mu\text{W}$  laser power; b PL spectrum for  $d = 50$  nm thick sample, at 5 mW laser power, measured on GaAs (black line) and on Au (red line) regions. The peak at 1.09 eV corresponds to the ground state and the peaks at higher values are due to the excited states



**Fig. 4** TRPL decay curves for  $d = 30$  nm thick sample, at 1 mW laser power, measured on GaAs (black line) and on Au (red line) regions. The measured IRF of the system is shown in light gray line



**Fig. 5** Lifetime data of the QD ground state extracted from TRPL decay curves for the samples for  $d = 10$  to 50 nm cap layer as a function of laser power. The results are given for the regions without (a) and with (b) the Au film at the ground state

(120 W/cm<sup>2</sup>), where the signal was high enough. The first excited state does not play an important role in the decay mechanism, as can be seen in Fig. 5a where the lifetime of the ground state is almost constant in the laser power range used in our experiments. The same argument is true also for the region where the Au film is present.

The decrement in the lifetime of the ground state can be explained by the coupling with the SPPs supported by the metallic film [15]. The important decay channels are the rate of exciton recombination with the creation of a photon  $\Gamma_{\text{rad}}$  (radiative channel), and into the GaAs surface states  $\Gamma_{\text{GaAs}}$  and SPPs  $\Gamma_{\text{SPP}}$  which constitutes a nonradiative channel, since the SPPs are free to propagate along the film they are not detected by our PL system. The total decay rate  $\Gamma_T$  for the exciton is given by

$$\Gamma_T = \Gamma_{\text{rad}} + \Gamma_{\text{GaAs}} + \Gamma_{\text{SPP}}, \tag{1}$$

where the decay rate into SPP modes is given by the Fermi's golden rule

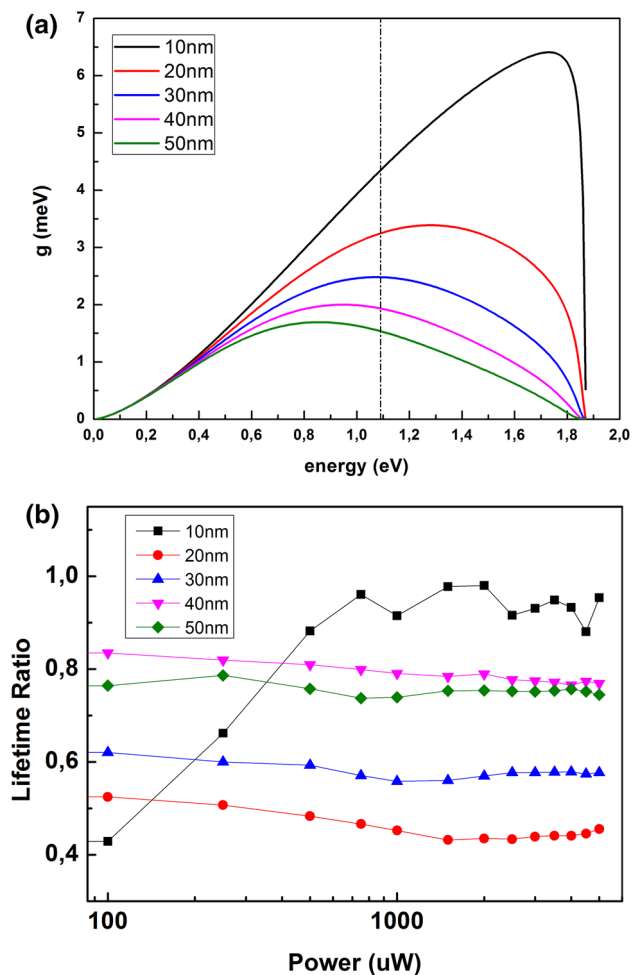
$$\Gamma_{\text{SPP}} = \frac{2\pi}{\hbar} g^2(\Delta) \rho(\hbar\omega = \Delta). \tag{2}$$

In Eq. 2,  $\rho(\varepsilon)$  is the local density of states of SPPs in the metallic film while  $g(\Delta)$  is the coupling factor between SPPs and the exciton ground state in the QD. The ground state energy of the QD is assumed to be  $\Delta$ . We obtained the coupling factor using an approach analogous to that given in [10]; in Fig. 6a, the result obtained for each cap layer thickness  $d$  is plotted as a function of the quantum emitter; the vertical dashed line is a guide to the eye for the QD ground state energy.

The competition between the SPP and surface states mechanisms is more evident for a sample with  $d = 10$  nm. However, for the samples with  $d = 20$ –50 nm the exciton relaxation is not very sensitive to this mechanism. For these samples, we see that as the cap layer increases the ratio in the lifetime at the regions with and without Au film, as shown in Fig. 6b, approaches a constant value. It is a strong evidence that SPPs play a major role in the recombination mechanism of the excitons in the QDs. This is more pronounced in the sample with  $d = 20$  nm than in the lifetime decay for  $d = 50$  nm, as expected by the decrement of the evanescent electromagnetic field of the SPPs and expressed as a decrease in the coupling factor shown in Fig. 6a.

### 3 Conclusion

We have shown an easy way to probe the interaction of excitons confined in a QD with SPP modes supported by a thin metallic film of Au. The main result is that the lifetime of the ground state decreases when compared to a sample



**Fig. 6** **a** SPP-QD coupling factor as a function of QD energy. The vertical dashed line is a guide to the eye for the QD energy in our experiments. **b** Ratio between the lifetime of QD ground state in the region with the Au film and without the Au film. The straight lines connecting the points are to guided the eyes

without the Au film. As expected by the theory that indicates an evanescent SPP electromagnetic field into the region where the QDs are placed, the influence of SPP on the lifetime decreases as the distance between the QDs and metallic film increases. We also observed a decrease in the exciton PL signal which is due to the excitation of the guided modes; then, this same approach may be useful for the design of an optical device which couples light to SPP without the use of plasmonic antennas and at a much smaller linewidth. Better linewidth precision of the SPPs could be obtained by using QWs instead of QDs. Another result that we have not discussed here is that the ratio between the first excited state to ground state PL has a strong modification when we compare the samples with and without the Au film. This shows that the coupling to first excited state is stronger than to the ground state. The lifetime of these states was not investigated since it is

below the limits of our equipments; however, this opens up a path for future investigations.

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