Nanoscale Systems for Optical Quantum Technologies

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D3.2 Plasmon launching

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Version history

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Deliverable Description

This deliverable corresponds to the aim of demonstrating **efficient plasmon launching in hybrid devices containing erbium ions and graphene**. Here, optically excited erbium ions, rather than decaying radiatively to far-field light (emitted photons), will decay through an alternative channel, where plasmon-polaritons are launched in a nearby graphene sheet. This project is mainly carried out by CNRS-CP (growth of thin films) and ICFO-NOE (fabrication of erbium-graphene hybrid devices and optoelectronic measurements), and builds upon the foundational work of these two groups, where erbium-induced plasmon launching was first demonstrated [Tielrooij2015]. By significantly reducing the thickness of the erbium layer compared to this previous work, more efficient plasmon launching is expected, where the aim is to reach a **plasmon-induced Purcell factor of >10.** This plasmon-induced Purcell factor corresponds to the relative reduction of photon emission of a film of erbium emitters due to the launching of graphene plasmons. Because of some unforeseen difficulties, we have not fully reached this goal. We will present various strategies to overcome the current challenges.

Concept

The properties of a light emitter depend on its dielectric environment. Graphene can act as an extreme kind of dielectric environment that leads to highly efficient non-radiative energy transfer from the emitter to graphene. As a result, the emission will be quenched and its decay rate will be enhanced. There are three distinct regimes of emitter-graphene coupling, depending on the carrier density, or equivalently, the Fermi energy E_F, of the graphene sheet. At low carrier density ($E_F < 0.5E_{em}$; with E_{em} the energy of the emitted photons – 0.8 eV in the case of Er^{3+} emitters), energy transfer leads to electron-hole pair creation, and at high carrier density ($E_F > 0.7E_{em}$), to launching of graphene plasmon-polaritons. At intermediate carrier densities ($0.5E_{em} < E_F < 0.7E_{em}$) energy transfer is less efficient due to Pauli blocking.

Efficient energy transfer from erbium emitters to graphene requires that the distance between the erbium emitters and the graphene sheet is small, significantly sub-wavelength. Typical hybrid erbium-graphene devices thus consist of a thin layer of erbium-doped Y_2O_3 (purple layer in figure on the right) with thickness t, on top of a silicon substrate. Then on top of the erbium layer, there is a monolayer graphene sheet (grey area in the figure on the right) grown by chemical vapour deposition (CVD), and finally there is a polymer electrolyte on top of the graphene for controlling its Fermi energy. Typically, the graphene is patterned into a Hall bar with 6 metal contacts. Two additional metal contacts are in electrical contact with the polymer electrolyte and used as gates.

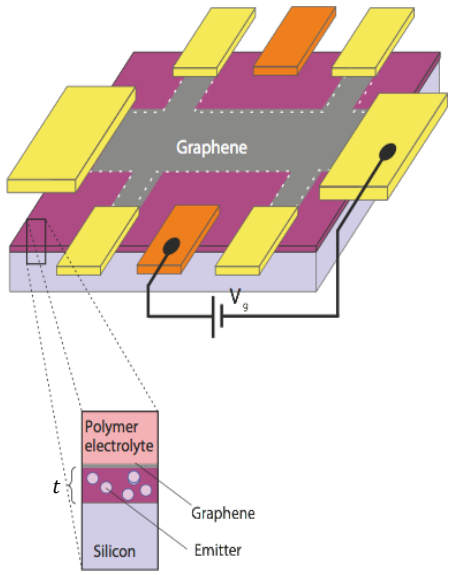

Fig. 1: Typical layout of erbium-graphene devices.

Numerical simulations

We have performed numerical simulations to determine what kind of *plasmon-induced Purcell factor* and emission contrast we can achieve, and in particular how thin the erbium-containing substrate layers should be.

Purcell factor and emission contrast

The total decay rate of excited erbium ions separated by a distance d from a graphene sheet is given by

$$\Gamma_{\text{tot, g}}(d) = \Gamma_{\text{rad}} + \Gamma_{\text{loss}} + \Gamma_{\text{e-g}}(d)$$

where Γ_{rad} is the radiative decay rate of erbium ions in bulk substrate, Γ_{loss} represents the non-radiative loss rate (not due to graphene), and $\Gamma_{\text{e-g}}(d)$ is the coupling rate between erbium ions and graphene. The non-radiative loss rate is caused by impurities in the thin film or by surface defects, as we will discuss in more detail later (see section on erbium film characterization).

The enhanced decay rate due to erbium-graphene coupling leads to a decrease in emission. The emission F in the presence of graphene (subscript g) and without the presence of graphene (subscript 0) is given by

$$F_{\text{g}/0}(d) \propto \frac{\Gamma_{\text{rad}}}{1 + \frac{\Gamma_{\text{tot, g}/0}(d)}{P_{\text{exc}}\, \text{C}_{\text{exc}}}}$$

Here P_{exc} is the excitation power and C_{exc} is the excitation rate that describes the creation of excited state population in the erbium ions. The factor $\text{P}_{\text{exc}}\text{C}_{\text{exc}}$ becomes important when the excited state population starts saturating, such that the emission is no longer linearly increasing with excitation power P_{exc}. In the low-power regime, the emission contrast reduces to

$$\frac{F_0(d)}{F_{\text{g}}(d)} \approx \frac{\Gamma_{\text{tot, g}}(d)}{\Gamma_{\text{tot, 0}}(d)}$$

In order to obtain the emission contrast for an erbium film with thickness t, we integrate the emission over all distances within the film thickness

$$F_{\text{film, g}/0} = \int_0^{\text{t}} \partial d\, F_{\text{g}/0}(d)$$

We will now define the *graphene-induced, effective Purcell factor* of a hybrid erbium-graphene device as $\text{F}_{\text{film, 0}}/\text{F}_{\text{film, g}}$. With graphene tuned to the high-density regime, where plasmon launching occurs, this factor will correspond to the *plasmon-induced, effective Purcell factor*, where we aim for a factor 10.

Simulation results

We obtain the erbium-graphene coupling $\Gamma_{e\text{-}g}$ as a function of Fermi energy and ion-graphene distance *d* using the *"Etot"* code developed by Javier García de Abajo, which takes into account the complete dielectric environment of our hybrid devices. For other variables, we use experimental data as in the table below. We vary the loss rate over three orders of magnitude to obtain a good understanding of its effect.

Variable	Value	Method
$\boldsymbol{\Gamma}_{rad}$	$(8\ ms)^{-1}$	Lifetime measurement on bulk substrate of Er-doped Y_2O_3
$\boldsymbol{\Gamma}_{loss}$	$(2\ \mu s - 0.2\ ms)^{-1}$	Lifetime measurements of thin films of Er-doped Y_2O_3
$\mathbf{C}_{exc}\boldsymbol{P}_{exc}$	$(1\ ms)^{-1}$	Measurement of emission vs. excitation power

In Fig. 2 we show as a function of substrate thickness the emission contrast without/with graphene $F_{film,\,0}/F_{film,\,g}$ for two different graphene carrier densities – one corresponding to the plasmon regime and one to the regime where electron-hole pairs are created. The case on the left corresponds to the *plasmon-induced, effective Purcell factor*, as we have defined it. Clearly, in order to reach our goal, we would need to have a film thickness below 15 nm. It is also evident that with a loss rate of $(0.002\ ms)^{-1}$ it will be challenging to reach our goal even with a film as thin as 5 nm.

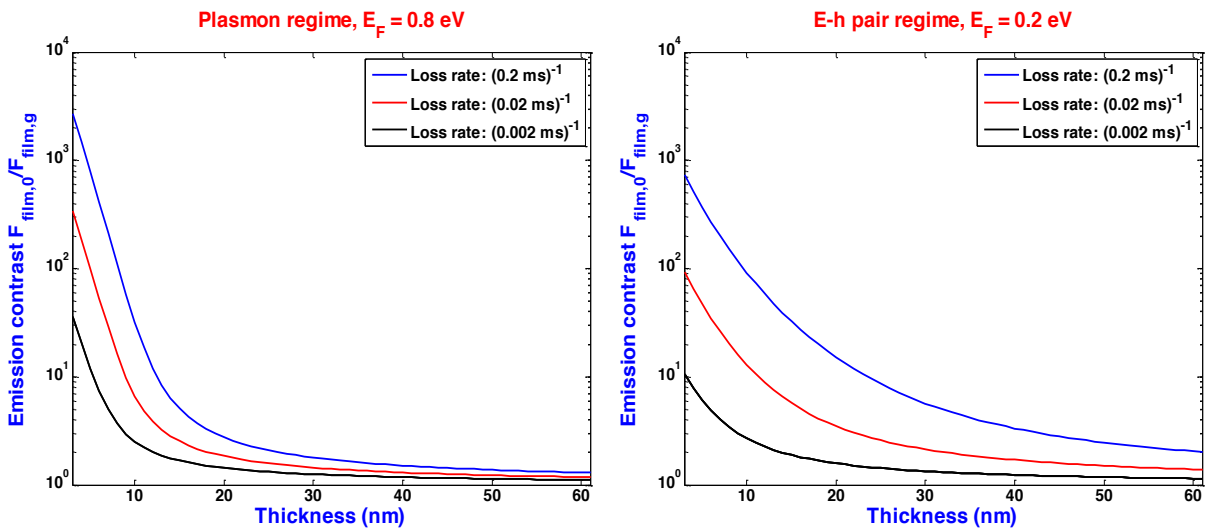

Fig. 2: Emission contrast as a function of thickness of the erbium layer in the plasmon regime (left) and electron-hole pair regime (right).

Experimental results

Erbium film characterization

The thin Er^{3+}:Y_2O_3 substrates were prepared by CNRS-CP using two different methods: Metal Organic Decomposition (MOD) [Andriamiadamanana2013], and Atomic Layer Deposition (ALD). The former technique was used to grow films with a thickness down to ~40 nm, whereas the latter technique was used to create even thinner films, down to ~5 nm. We characterize the emission lifetimes of these films by exciting with a temporally modulated 532 nm focused light beam and measuring erbium emission using a "Quantique" single photon detector (provided by ICFO-QP). We then use a "PicoHarp" to perform time-correlated photon counting with multistop mode, in order to generate lifetime histograms. We find that the thinner films show faster decay (see Fig. 3). This suggests that there is a loss channel present in these thin films, likely related to the surface effects and/or impurities in the film. The thickest film (300 nm) shows exponential decay with a lifetime of ~0.7 ms, which is ~10x shorter than erbium ions in bulk material, meaning that already in this film there is substantial loss. For thinner films, the decay is not exponential and contains at least one large very fast component. For film thicknesses of 6, 10, 11, and 40 nm, we find 1/e decay times of 0.005, 0.008, 0.025, and 0.15 ms, respectively.

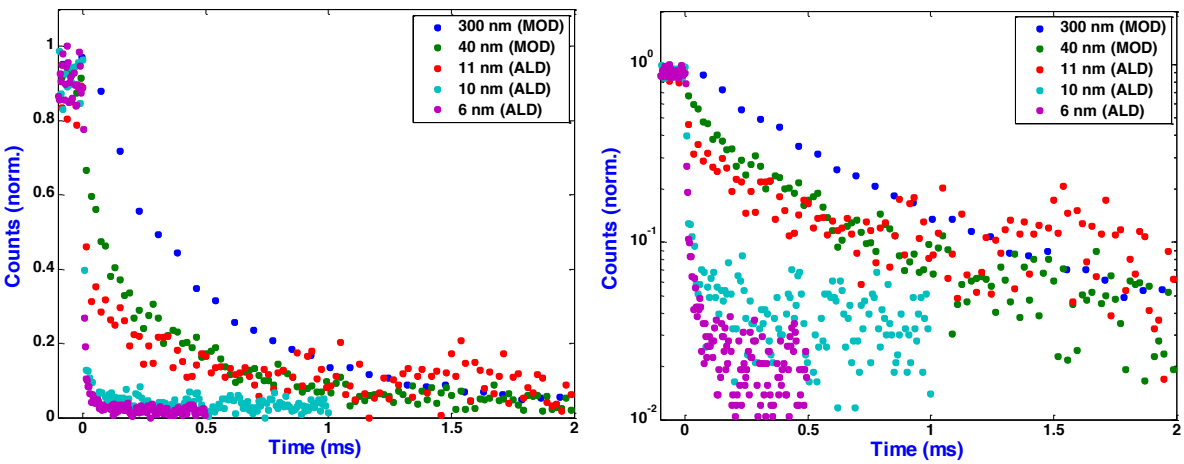

Fig. 3: Lifetime measurements of erbium thin films, comparing film thickness with linear (left) and logarithmic (right) vertical scale.

In order to reduce the loss – at least the loss induced by the surface – we applied a 1-2 nm thick capping layer of Al_2O_3 grown by ALD (see Fig. 4). Even though a large fast decay component is still present, the tail of the decay is – in some cases – slower, indicating reduced loss. However, even in the cases that the capping layer let to improvement, still >60% of the decay is completed within 0.01 ms. These film characterization results show that the main loss occurs with a rate that is between $(0.002\ \text{ms})^{-1}$ and $(0.02\ \text{ms})^{-1}$, corresponding to the black and red curves in our simulations results, respectively.

We will discuss a number of strategies to reduce the loss in the Outlook section. Furthermore, we note that due to the intrinsic loss in the erbium film, the emitters have a low quantum efficiency (QE). However, if the erbium-graphene coupling is much

stronger than the loss rate, $\Gamma_{e\text{-}g} \gg \Gamma_{loss}$, the quantum efficiency is enhanced due to emission into graphene plasmons.

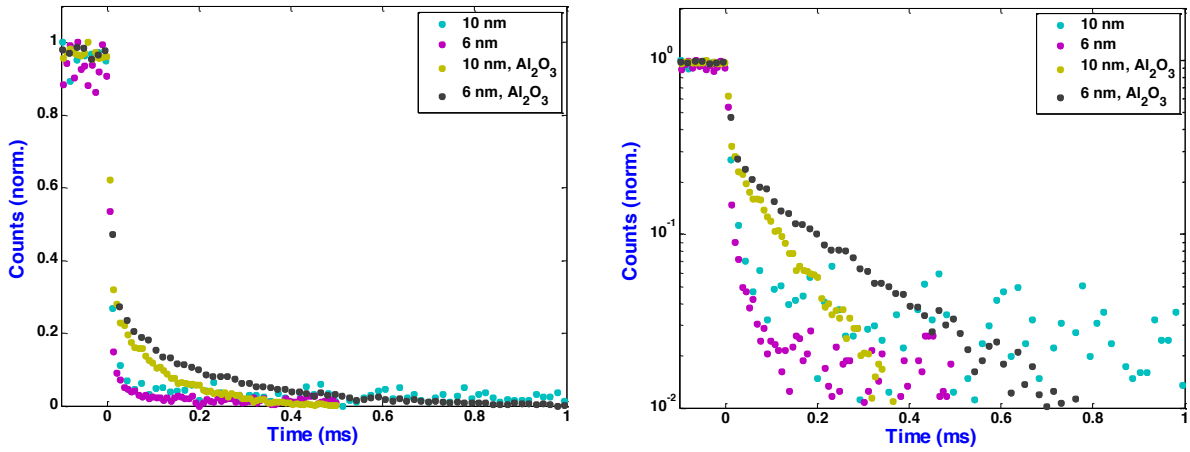

Fig. 4: Lifetime measurements of erbium films of 6 and 10 nm thickness with and without a 1-2 nm Al_2O_3 capping layer with linear (left) and logarithmic (right) vertical scale.

Characterization of hybrid devices

We have fabricated close to 20 different erbium-graphene devices as in Fig. 1, using around 9 different substrates with erbium layer thicknesses varying between 15 nm and 6 nm. Some substrates had an Al_2O_3 capping layer and some did not. In some cases, we added a plasma cleaning step (oxygen/nitrogen) before transferring the CVD graphene. For all these different devices, we observed a similar emission contrast of $F_{film,\,0}/F_{film,\,g} \sim$ 2-3, where the emitters usually decay by exciting electron-hole pairs in graphene. This regime is most easily accessible experimentally and has a similar contrast as the plasmon regime. Figure 5 shows two examples of emission images.

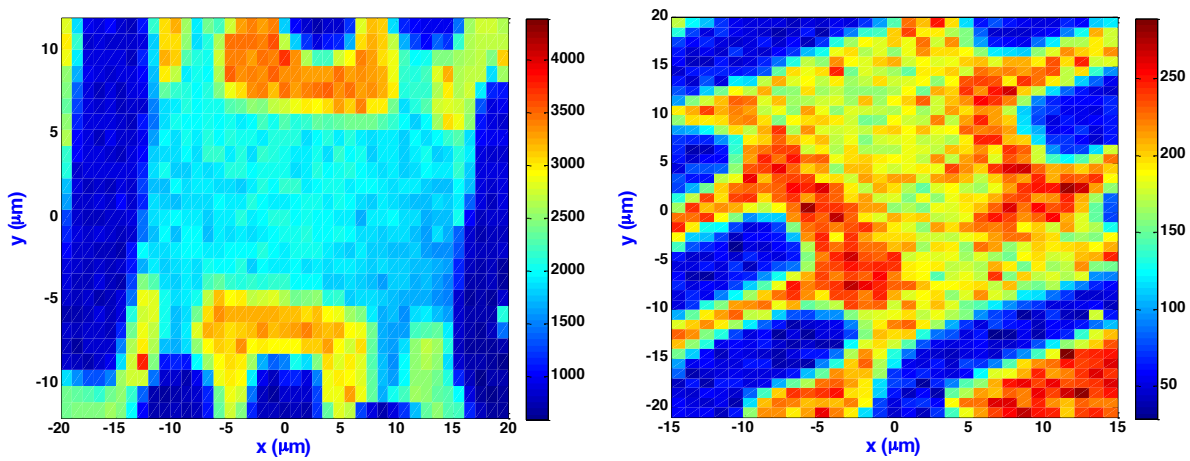

Fig. 5: Erbium emission images for graphene devices on erbium films with a thickness of 11 nm (left) and 6 nm (right). The color scale represents emission in counts/s.

The emission images show strongly reduced emission at the metal contacts (blue color), due to reflection of the excitation light. However, also on locations where graphene is present, there is reduced emission (cyan and green colors). This shows that there is significant coupling between erbium emitters and graphene in these thin films. This

coupling is actually very strong as it competes with the strong internal loss in the films. From the observation of graphene-induced emission quenching, we can conclude that the *graphene-induced, ideal Purcell factor*, which ignores loss in the thin film, is: $\Gamma_{e\text{-}g}/\Gamma_{rad}>10^3$. This is a highly encouraging result.

Finally, we compare lifetime traces with the excitation laser exciting a region without graphene and a region with graphene. We show a typical example of such a comparison for a device with erbium layer thickness of 15 nm in Fig. 6 (in the electron-hole pair excitation regime).

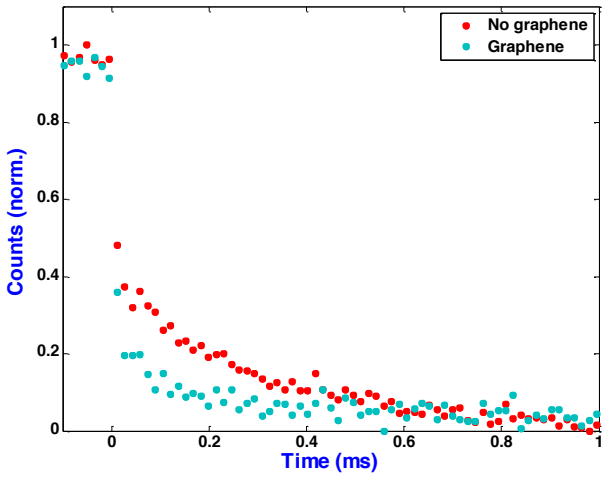

Fig. 6: Erbium lifetime trace with and without graphene.

Conclusion

We have succeeded in fabricating hybrid erbium-graphene devices (as in Fig. 1) with ultrathin erbium layer thicknesses, and in observing graphene-induced energy transfer from erbium to graphene, manifested in reduced emission (see Fig. 5) and faster decay (see Fig. 6). The emission contrast, and thus the *graphene-induced, effective Purcell factor*, we have obtained is below 10 and therefore not as high as we had aimed for. We suggest that the main reason for the relatively low contrast is the strong loss channel that occurs in these ultrathin films (see Fig. 3). Indeed, using a loss rate of $(0.002\ \text{ms})^{-1}$, the simulations predict a contrast of 2-4 in the electron-hole pair regime (see Fig. 2), similar to what we observe. Additionally, we suggest that the possible presence of (charged) impurities (perhaps during graphene transfer) in between the erbium ions and the graphene could screen the interaction and reduce the contrast and/or increase the erbium-graphene distance. However, the fact that we observe a non-negligible emission and lifetime contrast due to presence of the graphene film, despite the strong internal loss, is a promising result that shows that indeed emitter-graphene coupling can be very efficient, as illustrated by the *graphene-induced, ideal Purcell factor* $>10^3$.

Outlook

We suggest the following strategies to overcome the current challenges and reach the goal of a *graphene-induced, effective Purcell factor > 10*. In brackets we indicate the amount of time needed to produce conclusive results about these approaches.

- ⇨ Exfoliated graphene flake instead of CVD graphene (4 months)

By using an exfoliated graphene flake, we avoid possible contamination that could be introduced by wet graphene transfer onto the erbium film. This could lead to a reduced erbium-graphene distance and possibly to less screening of the erbium-graphene interaction by interfacial charges. Also, we can avoid additional fabrication steps that could affect the cleanliness of the erbium-film graphene interface.

- ⇨ Exploit other energy levels (6 months)

Finally, we suggest exploring different erbium transitions that could couple more strongly to graphene plasmons. In particular, we suggest using excitation light with a wavelength of ~980 nm to populate the $^4I_{11/2}$ level, where ions can relax to the $^4I_{13/2}$ level (a 3 μm transition) and then to the $^4I_{15/2}$ level (the 1.5 μm emission that we typically probe). The idea then is that plasmon-coupling enhances the decay rate of the 3 μm transition, making the overall decay faster.

- ⇨ Reduce substrate loss (9 months)

There are various ways of achieving this. First of all, we will try to find ways to minimize the amount of impurities that are introduced into the film during ALD. Secondly, we will optimize the cleaning of the films after its growth, where we have indications that annealing at elevated temperatures (>800°C) could be beneficial. Thirdly, we will keep the substrates in high vacuum (10^{-7} bar) before transferring the graphene and keep them in vacuum during measurements. Finally, we suggest to measure at lower temperatures, where loss should be reduced.

K.J. Tielrooij et al., Electrical control of optical emitter relaxation pathways enabled by graphene, Nat. Phys. 11, 281 (2015)

C. Andriamiadamanana et al., Erbium-doped yttria thin films prepared by metal organic decomposition for up-conversion, Thin Solid Films 537, 42 (2013)