Nanoscale Systems for Optical Quantum Technologies

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D3.10 Plasmon induced ion coupling

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Author(s):	Daniel Cano (ICFO-NOE)

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V1	17/09/2019	D. Cano (ICFO-NOE)	First draft
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Deliverable Description

In this deliverable, we report on the experimental development of hybrid erbium/graphene devices that integrate the main functionalities required for plasmon-mediated ion-ion coupling. In particular, our devices have strong erbium-plasmon interactions that can be modulated at high frequencies by applied gate voltages. This fast temporal control over the erbium-plasmon interactions has been regarded as a potential way to generate ion-ion coupling [Man12].

In the theoretical part, we have investigated the signatures of plasmon-mediated ion-ion coupling. The most relevant signatures are the collective emission and the suppression/enhancement of spontaneous decay. These findings can be used for new proposals and novel experiments with our hybrid erbium/graphene devices. Unfortunately, we have not been able to observe these signatures experimentally given the technical challenges of controlling these interactions at nanoscale distances near a dissipative medium like graphene.

Both experimental and theoretical research activities of this deliverable are connected with several tasks of the NanOQTech project. The experimental development of hybrid erbium/graphene devices combines the technology of erbium-doped thin films with optimized optical properties (task 1.2) and the fabrication of graphene devices with controlled plasmon launching and rapid tunability of the erbium-plasmon interactions (task 3.1 and 3.2). The theoretical work and proposals for new experiments were carried out as described in task 3.5.

Introduction

The control of the coherent interactions between quantum systems is at the heart of quantum technologies. In particular, the control of the interactions that are based on the exchange of photons can be achieved by means of photonic cavities, which are used to enhance and modulate the coupling of the quantum systems to the electromagnetic fields. The experimental realizations of cavity-mediated interactions between ultra-cold atoms [Bau10, Nor18] and between superconducting qubits [Axl18] have led to the observation of intriguing phenomena, including new quantum phase transitions and long-distance entanglement. Despite of these great advances, similar technologies have not yet been implemented efficiently in solid-state quantum systems that can be interconnected in nanoscale opto-electronic devices, with which quantum technologies can go to real applications.

The ambition of this deliverable is to show that graphene plasmons can mediate the interactions between rare-earth ions, in a similar way as photonic cavities mediate the interactions between cold atoms or between superconducting qubits. The advantage of graphene is its capability to strongly confine the electromagnetic field in the form of plasmon polaritons, whose wavelengths are two orders of magnitude smaller than those of free-space photons of the same frequencies. This strong confinement provides an excellent platform for strong interactions.

A major drawback of graphene plasmonic cavities is their low-quality factors. Although they offer extraordinarily high Purcell factors thanks to their strong electromagnetic confinement, their quality factors are typically low, i.e. plasmon lifetimes are small as compared to photonic cavities. Graphene plasmons decay with rates of the order of 1 THz, whereas the spontaneous decay rate of erbium ions is only 100 Hz. Therefore, Purcell factors higher than 10^{10} are required to make the plasmon-mediated information transfer between ions faster than the rapid decoherence of graphene plasmons. Unfortunately, such extremely strong erbium-plasmon coupling is very difficult.

To overcome the low-quality factors of graphene, novel methods to create plasmon-mediated ion-ion interactions have been proposed [Man12, Veg16, Zha19a, Zha19b, Zha19c]. Instead of using the cavity as a bus that transfers photons or plasmons deterministically between different ions, graphene is used as a dissipative medium that enhances the collective spontaneous decay of the ions by plasmon emission. In this manner, by engineering the collective decay of the ions, it is possible to generate ion-ion interactions and collective non-classical states. Because this method is based on dissipation, it does not require high quality factors, and graphene plasmonic cavities can be used. On the other hand, one important requirement is to have temporal control of the collective spontaneous decay, which has to be modulated at higher frequencies than the decay rate of the ions [Man12, Veg16]. In this deliverable, we demonstrate experimentally that this requirement can be fulfilled in the hybrid erbium-graphene devices that we developed and characterized during the period of the project.
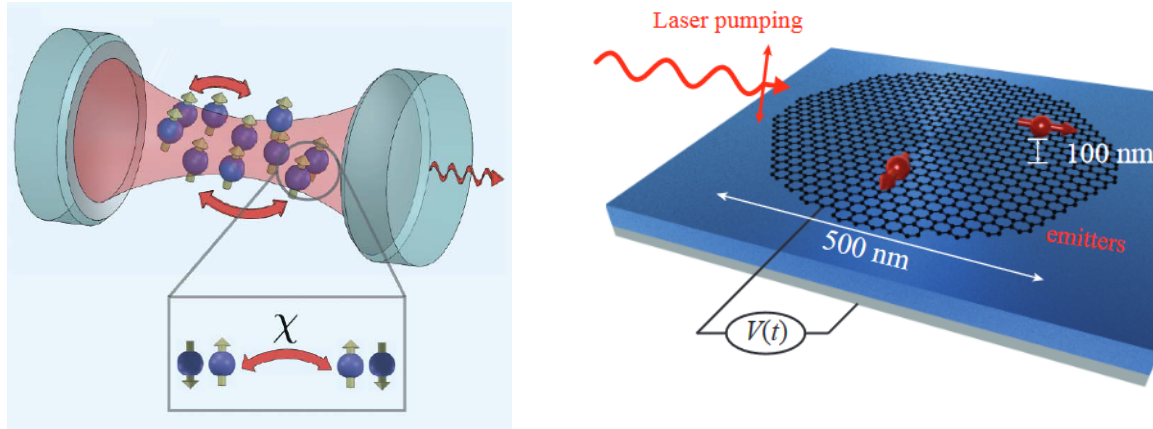
Figure 1: (Left) Cavity-mediated atom-atom interactions: a cavity mediates spin-exchange interactions in which one atom emits a photon into the cavity that is then absorbed by another atom, driving anticorrelated spin flips. **(Right) Plasmon-mediated ion-ion interactions:** two ions are excited and their collective spontaneous decay is modulated electrically through the plasmons of a neighboring graphene disk. (The figures have been taken from Refs. [Nor18, Man12]).

Optoelectronic devices for the temporal control of erbium-plasmon interactions

Our experimental efforts have been focused on the development of hybrid erbium/graphene devices that integrate the functionalities required for plasmon-mediated ion-ion coupling and non-classical correlations. In particular, we have created devices with strong erbium-plasmon interactions and very fast electro-optical modulation, up to 5 kHz frequencies (see Figure 2). In these devices, graphene acts as an extreme kind of tuneable dielectric environment that cause highly efficient non-radiative energy transfer from the excited erbium ions. The energy transfer is described by the Purcell factor, which can be modulated at high frequencies by modulating the carrier density of graphene. The starting point for this report is the result described in the previous deliverable (3.2), where we already showed high Purcell factors. The development of the devices has implied intensive collaboration between the CNRS-CP (growth of erbium-doped films) and ICFO-NOE (fabrication of erbium/graphene devices and optoelectronic measurements) partners.

High Purcell factors

The experimental work to achieve high Purcell factors is described in tasks 1.2 and 3.1 of the project. High Purcell factors require that the distances between erbium and graphene are shorter than plasmon wavelengths ($\sim$10 nm). For this purpose, the decisive step of our work was to optimize the growth and the thermal post processing of very thin films (only 10 nm) of erbium-doped yttria, obtaining high purity and good crystallinity as well as almost pure radiative decay. This optimization was essential since nano-sized erbium-doped crystals commonly suffer from intrinsic non-radiative decay rates that would compete against the interactions with graphene plasmons. The optimization process was realized at CNRS-CP, and the results were described in deliverable D1.4.

Fast modulation of the erbium-plasmon interactions

We show that we can reach modulation frequencies of the erbium-graphene interactions higher than the decay rates of most of the ions. This is described in tasks 3.1 and 3.2 of the project, and is a prerequisite for temporal quantum control and ion-ion coupling [Man12]. Our devices integrate two kinds of gates that act together. First, there is a top gate, which consists of a transparent solid polymer electrolyte that is placed on top of graphene and which can provide high Fermi energies up to 1 eV. This top gate is used to reach the plasmon regime, although it cannot be modulated at high frequencies due to its high electrical resistance. For the fast modulation, the devices also include a back gate connected to the back side. A SiO_2 layer electrically isolates the back gate from graphene and the other electrodes.
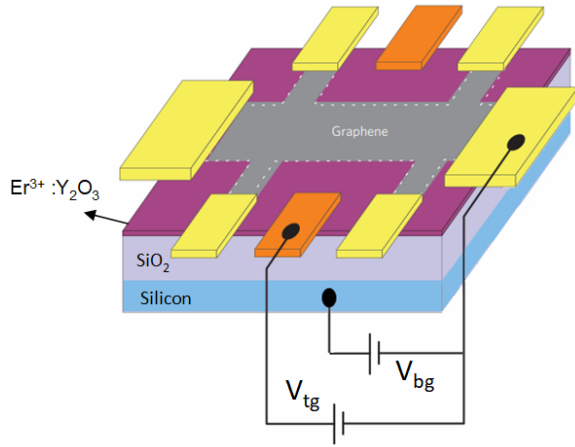
Figure 2: A Hall-bar shaped sheet of graphene is placed directly on top of the erbium-doped film. The top- and back-gate voltages are used to tune the carrier density in graphene – with V_{tg} we achieve high carrier densities and V_{bg} is used for fast modulation.

Measurements of high Purcell factors induced by graphene plasmons

The generation of graphene plasmons requires high carrier densities. It has been shown that graphene behaves as a plasmonic material when the Fermi energy is $|E_F| > 0.7\ E_{em}$, where E_{em} the energy of the emitted photons ($E_{em} = 0.8$ eV in the case of Er^{3+} emitters). Figure 3 (left) shows the measurements of erbium emission as a function of Fermi energy. The plasmon regime can be recognized by the decreasing slope in erbium emission at high Fermi energies. In these measurements, we excite the ions with a 532 nm focused laser spot and measure their emission at 1.54 µm using a home-built scanning confocal microscope setup [Tie15].

To quantify the coupling of the erbium ions to the graphene plasmons, we do time-resolved fluorescence measurements. For this purpose, the excitation laser intensity is modulated into square pulses by switching on and off the signal of an acousto-optic modulator. Figure 3 shows the decay curves on different regions of the device, with and without graphene, denoted as $n_{on}(t)$ and $n_{off}(t)$ respectively. The decay curve on graphene is highly multi-exponential, especially at the beginning of the decay, as a consequence of the strong dependence of the erbium-graphene interactions on the distance. If we assume that every ion decays exponentially with a specific relaxation rate γ, the decay curve for the whole distribution of ions will be

$$n_{on}(t) = \int_0^\infty \frac{P_{on}(\gamma)}{\gamma} e^{-\gamma t} d\gamma\,, \qquad n_{off}(t) = \int_0^\infty \frac{P_{off}(\gamma)}{\gamma} e^{-\gamma t} d\gamma$$

where $P_{on}(\gamma)$ and $P_{off}(\gamma)$ are the probability distributions with and without graphene, respectively. Here, we have considered that the emission of every ion is inversely proportional to its decay rate because the excitation laser intensity is much lower than the saturation intensity in our experiments. The decay rate has four contributions,

$$\gamma = \gamma_e + \gamma_m + \gamma_{nr} + \gamma_{gr}\,,$$

which represent, in that order, the electric and magnetic dipole moments of the ion transition, the intrinsic non-radiative decay channels of the erbium-doped film, and the

energy transfer from the ions into graphene plasmons. Only the electric dipole moment of the transition interacts with the graphene plasmonic modes – these are always transversal magnetic. Therefore, we can write

$$\gamma_{gr} = (F_P - 1)\gamma_e,$$

where F_P is the Purcell factor, which strongly depends on the ion-graphene distance. To obtain the probability distribution $P(\gamma_{gr})$ of decay into graphene modes, we have used the following relation:

$$P_{on}(\gamma) = \int_0^\gamma P_{off}(\gamma - \gamma_{gr})P(\gamma_{gr})d\gamma_{gr},$$

By doing the deconvolution of $P_{on}(\gamma)$ and $P_{off}(\gamma)$, we obtain $P(\gamma_{gr})$. We represent the distribution $P(\gamma_{nr})$ in Figure 4. At least 55% of the ions have decay rates into graphene higher than 10^3 s^{-1}, i.e, Purcell factors higher than 10. This is the milestone MS3 of WP3 as well as an important step of the realization of task 3.1.
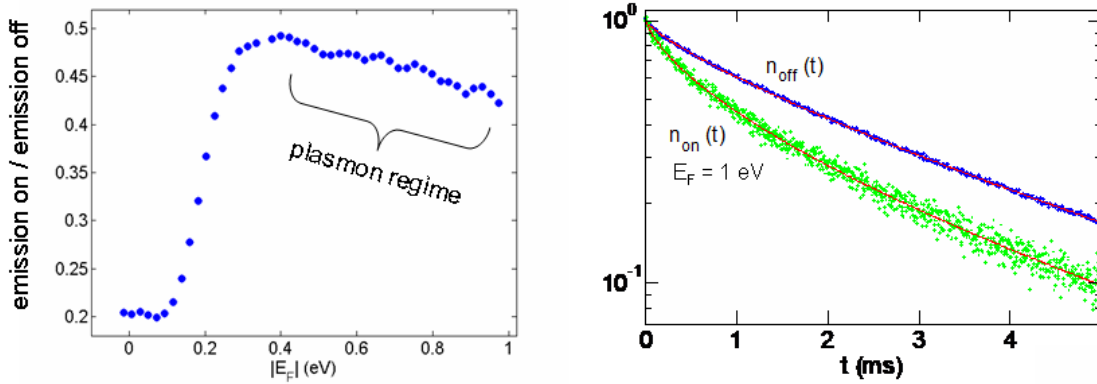

Figure 3: (Left) Photon emission on graphene, normalized to the emission of the bare erbium substrate, as a function of the Fermi energy of graphene. At low Fermi energies, the emission is low due to the non-radiative energy transfer into electron-hole pair creation. As the carrier density increases, the electron-hole pair becomes forbidden, which results in an increase in erbium emission. For higher Fermi energies, the increasing role of graphene plasmons results in a decreasing emission. **(Right)** Decay curves of Erbium ions measured on two different regions, with and without graphene. Measurements on graphene have been done for a Fermi energy of 1 eV.
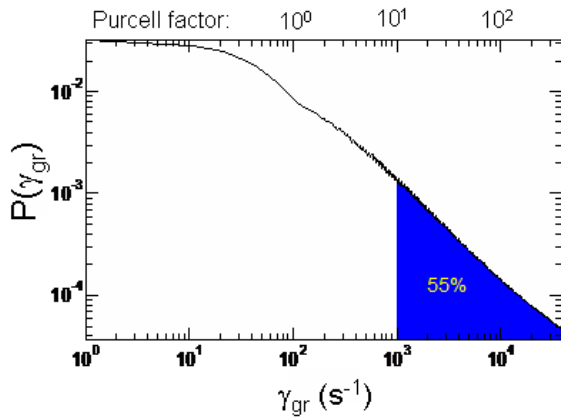
Figure 4: Probability distribution of the spontaneous decay rates (and Purcell factors) induced by graphene plasmons. More than half of the ions have Purcell factors larger than 10. This probability distribution is normalized by considering only the values of γ_{nr} smaller than 10^5 s^{-1} because higher decay rates are not detectable with our experimental setup.

Measurements of fast temporal control over erbium-plasmon interactions

To show that we can reach modulation frequencies higher than the decay rates γ_{gr}, we apply a time-dependent gate voltage that creates an oscillation in the Fermi energy of 0.2 eV amplitude superposed to a constant bias Fermi energy of 0.7 eV. The modulation of the plasmon local density of states results in a modulation of the erbium emission, as shown in Figure 5. For modulation frequencies above the decay rates γ_{gr}, the maximum and minimum of the oscillating emission get shifted and the amplitude gets smaller. We model this oscillation with rate equations [Foo05], considering the time-dependent decay rates given by the corresponding Purcell factors of the oscillating plasmon density of states [Tie15].
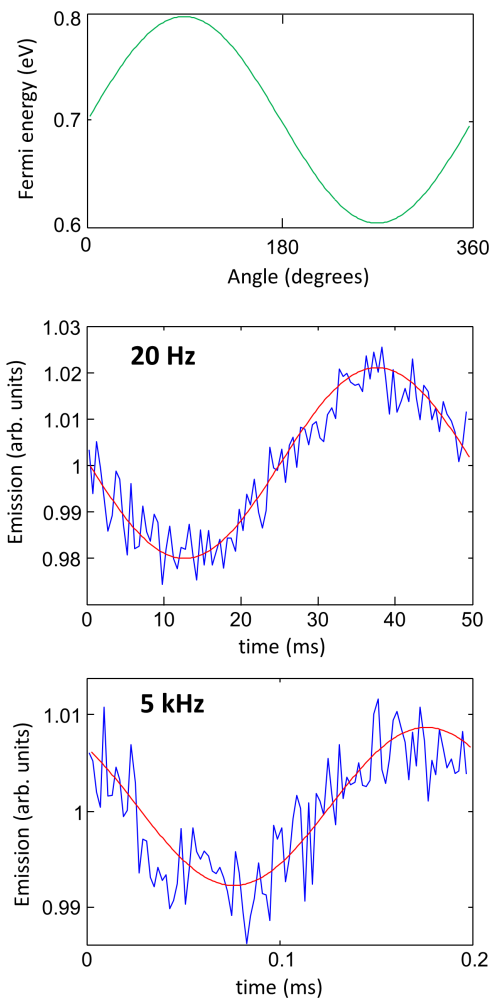

Figure 5: (Top) Oscillation of the Fermi energy done by modulating the back gate voltage. **(Middle and bottom)** Measurements of time-resolved erbium emission while the Fermi energy is modulated, for two different modulation frequencies: 20 Hz and 5 kHz. For these measurements, the photons are detected with a photon counter during many oscillation periods. The obtained emission signals are divided by their mean value. The red lines show the simulations of the rate equations assuming a time-dependent decay rate. The maximum and minimum of the emission signal at 5 kHz are shifted with respect to the emission at 20 Hz, which is evidence that the modulation frequency is higher than the decay rate of a large portion of the ions.

Theoretical work on the signatures of plasmon-mediated ion-ion coupling

We developed theoretical models to analyse the consequences of plasmon-mediated ion-ion coupling and to design new experiments in our hybrid erbium/graphene devices. This work has been realized by the partner AU in relation to the activities of task 3.5 of the

project. We find that plasmon-mediated ion-ion coupling leads to intriguing phenomena like quantum correlations, and superradiant and subradiant decay via plasmon emission.

We have modelled the launching of single and coherent plasmon pulses from an excited ensemble of ion emitters. To maintain excitation in the ensemble, the excitation proceeds via so-called timed-Dicke states, with spatial phases that are ultimately phase matched to the plasmon wavenumber. By a sequence of Raman pulses, it is thus possible to "pump" the wavenumber to match any desired value, only after the last pulse and monitor the subsequent emission. The quantized emission is subject to dispersion and dissipation, and we find that the plasmon generation is highly directional and described by a general non-Markovian theory, offering a rate process limit or by an oscillatory regime, separated roughly by the constraint that the coherence length is traversed by the plasmon pulse faster, slower than the collective emission time scale [Zha19a].
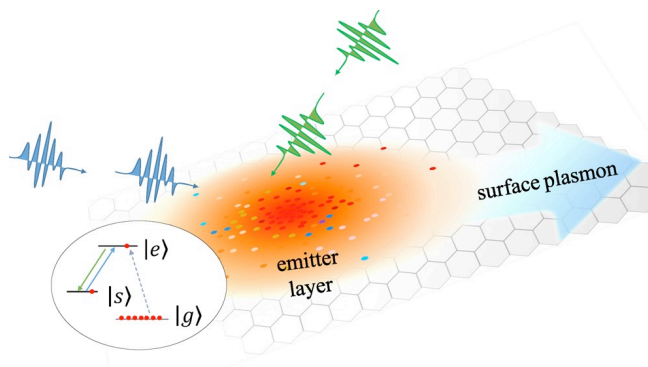
Figure 6: (From [Zha19a]) Raman pulses create spatially dependent phase coherence of emitters in a layer above a graphene surface. When the transition energy and spatial wavenumber matches the plasmon dispersion relation, a surface plasmon is launched.

We considered the specific case of graphene plasmon, described by the Drude model for our numerical analysis in [Zha19a], but in subsequent work we have been studying the prospects to observe amplification and plasmon lasing with other low-loss materials than graphene (work in progress).

Our work on collective emission and the suppression/enhancement of decay from the timed-Dicke states has been followed by general analyses of the subradiance and superradiance properties of regular spatial structures of quantum emitters. For a 1D chain of N equidistant emitters we have analytically demonstrated the existence of subradiant states with lifetimes scaling as N^3/m^2, where m =1,2, … enumerates the states. These results were so far only hinted at from numerical studies. Our analysis also explains the existence of states with more excitations of a "fermionic nature" and decay lifetimes scaling as $N^2/m^2 + N^3/n^2$, where n and m are different [Zha19b]. Among the multiple excited states, we have furthermore identified "dimer states", i.e., very long lived excited states of the chain of atoms, with the excitations being always close to each other, but with a fully spread out "center of mass". These states are even longer lived than the singly excited states of the string [Zha19c]. The latter findings may have consequences for the use of ions as emitters in nanocrystals for long memory storage, if we can overcome inhomogeneous broadening effects.

Conclusion: prospects of plasmon-induced ion-ion interactions

The devices realized for this project integrate the basic functionalities required for plasmon-mediated ion-ion interactions as described in published proposals [Man12] and in the theoretical analyses given by task 3.5 [Zha19a, Zha19b, Zha19c]. In particular, any desired spontaneous decay profile can be created, exploiting the fast tunability of graphene plasmons and their strong interactions with the neighbouring erbium ions. This temporal control over the erbium ion dynamics is achieved by simply applying a variable gate voltage using conventional electronics and following the requirements identified in tasks 3.1 and 3.2. In terms of stability and robustness, this way to tune the cavity by purely electronic means is an important advantage over the nano positioning systems and motion controls required to tune the optical cavities with mirrors.

Nonetheless, before the observation of plasmon-mediated ion-ion interactions, a few implementations have still to occur. The heterogeneous spectral broadening of the erbium ions represents a challenge because it reduces the number of ions that couple to the same plasmonic mode. This can be compensated by increasing the plasmon propagation lengths up to units of microns, thus covering a larger volume of the ion ensemble. For that purpose, we will use boron-nitride-encapsulated graphene made by exfoliation instead of the CVD graphene that we have used in our devices. Boron-nitride-encapsulated graphene made by exfoliation is the method to produce graphene with the highest carrier mobilities [Kre14], which results in longer plasmon propagation lengths. We used CVD graphene because of its low cost and ease of use, which is very convenient for optimization tests with large amounts of different devices. Another requirement for longer propagation lengths is low temperature, which implies the use of an experimental system with a cryostat. Longer propagation lengths of units of microns are very desirable to couple the highest possible number of ion emitters into the same plasmonic mode and to observe the collective effects of ion-ion coupling.

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