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Electron beam interrogation and control of ultrafast plexcitonic dynamics

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Abstract

In the strong coupling regime, photons and excitons exchange energy very rapidly such that they blend together, forming hybrid plexcitonic states. Recently, strong coupling of plasmons with a single fluorescent molecule was realized for the first time at room temperature using plasmonic nanocavities. Here, we show that electron beam excitation of a plasmonic nanocavity allows us to dynamically control the light-matter interaction forming the plexciton without changing the plasmonic nanocavity. Unlike dark-field microscopy, an electron beam has the ability to excite dark plasmons which, due to their non-radiative nature, allow for more energy cycles between the plasmon and the emitter to survive in a very lossy system. Directly comparing effective 2D (nanowire) and 3D (nanosphere) plasmonic nanocavities we demonstrate that the dark modes inherently present at lower frequencies in the nanowire-nanocavity can be used as a means to extend the quantum coherence between the plasmon and emitter, [opening the way towards dynamic tuning](#) of the strong-coupling dynamics via the electron

velocity. This opens new routes for dynamically addressing and controlling quantum emitters in the strong coupling regime via (the velocity of) the electrons.

Keywords

strong coupling, electron beam, quantum emitters, quantum dynamics, plasmonic nanocavities

Introduction

Nanoplasmonic devices have the ability to confine light in extremely small volumes¹⁻³ and enhance electromagnetic fields by several orders of magnitude. These properties have led to an emergence of exciting applications in colorimetric sensing,⁴⁻⁶ single molecule surface-enhanced Raman scattering⁷ and plasmonic lasers.⁸ Placing a quantum emitter (QE) such as a quantum dot or a dye molecule in [plasmonically enhanced fields](#), one can alter the local density of states to an unprecedented degree.^{9,10} Recently, advances in nanofabrication and excitation techniques have enabled us to reach the single-emitter strong coupling regime at ambient conditions using nanoplasmonic cavities and resonators¹¹⁻¹⁵ where the energy exchange between the quantum emitter and plasmon is much faster than the decoherence processes in the system.^{14,16} Hence, the QE exciton blends with the plasmonic mode, creating hybrid plexcitonic states which are part light, part matter. This cyclic energy exchange between the plasmon and exciton is characterized by Rabi oscillations observed for systems in the strong-coupling regime, which provide the characteristic anti-crossing behavior of far-field spectra commonly measured experimentally.

Concurrently, electron energy loss spectroscopy (EELS)¹⁷ has matured to become one of the most important experimental methodologies for characterizing nanoplasmonic environments.¹⁸⁻²² The EELS technique employs an electron beam passing at a very close proximity to a plasmonic structure, exciting its photonic modes and therefore transferring part of the

electrons' energy to the plasmons.^{23–26} Since the excitation of the plasmons occurs in the near-field, EELS has the advantage of exciting next to bright plasmons and also dark plasmons which cannot be accessed via typical optical techniques, such as dark-field microscopy. EELS also provides sub-nanometer spatial resolution and meV energy accurate results. Not surprisingly, this has established EELS as one of the most significant experimental tools for investigating plasmonic nanostructures. While many recent breakthroughs in nanoplasmonics were facilitated by EELS,^{27–29} the technique itself has been advanced to also map phononic states¹⁸ in the infra-red regime and the spatially resolved identification of isotopically labeled molecules.³⁰ Recently, the versatility of the method was proven in showing the strong coupling of emitters and plasmons excited by fast electrons in diverse geometries such as plasmon/exciton coupling between metal triangular particles and 2D materials³¹ or quantum dots placed in triangular bowtie antennae³² as well as plasmon/phonon coupling between 2D semiconductors and silver nanoantennae.³³ The experimental investigations have also been complemented by recent theoretical studies proposing more rigorous ways to describe the coupling between quantum emitters and plasmonic cavity modes using a full quantum description.^{34,35}

In this article, we propose to use an electron beam to dynamically tune and interrogate the strong coupling dynamics of a QE in a plasmonic nanocavity. We exploit the dark modes of plasmonic nanocavities and use the electron beam to efficiently excite both bright and dark plasmons. While the dark plasmons allow for a longer lived coherence (i.e. energy exchange with QEs), the electron beam velocity dictates the strength of the dark mode excitation, and hence the strong coupling dynamics with the QE. We start by considering a simple isolated spherical nanoparticle, for which well established analytical descriptions already exist,^{25,26} and identify the dependence of the dark and bright plasmon excitation on the electron velocity. We then move to plasmonic nanocavities, focusing on the 2D nanowire on mirror (NWoM) and the 3D nanoparticle on mirror (NPoM) configuration, where single molecule strong coupling with plasmons was recently realized experimentally at ambient conditions.¹³

2D plasmonic structures support dark plasmons spectrally degenerate with bright modes, while 3D plasmonic structures support spectrally isolated bright modes.³⁶ Utilizing this distinct property between the two plasmonic nanocavities, we show tunable Rabi oscillations with the electron beam velocity that, interestingly, manifest themselves differently in the near- and far-field spectra usually measured experimentally. This work paves the route to an improved understanding of the complex interaction between an emitter and the many plasmon modes present in plasmonic devices, and ultimately design future experiments³⁴ that make use of their unique properties.

Results

To describe how travelling electrons passing at a close proximity to a nano-metallic structure couple energy into the plasmonic modes, we **first focus on** an analytical description:²⁵ An electron beam passing at close proximity to a nano-metallic structure couples energy into both bright and dark plasmon modes. To understand the excitation strength of these bright and dark plasmon modes, we employ the analytical description developed by Garcia de Abajo.²⁵ An electron moving with velocity v produces an electric field which causes collective electron oscillations in the metal nanostructure placed in the vicinity of its trajectory (i.e. plasmonic excitations). In response, the plasmon modes induce a field $\mathbf{E}^{ind}$ that acts back on the electron, exerting a force that produces an energy loss.³⁷ For an electron moving with constant velocity $\mathbf{v}$ along a straight trajectory $\mathbf{r} = \mathbf{r}_t$, the total energy loss relates to the induced field $\mathbf{E}^{ind}$ via^{17,26}

$$\Delta E = e \int \mathbf{v} \cdot \mathbf{E}^{ind}[\mathbf{r}_e(t), t] dt = \int_0^\infty \hbar \omega \Gamma_{EELS}(\omega) d\omega, \quad (1)$$

from which we can extract the electron energy loss probability per unit of transferred frequency ω (i.e. spectra):

$$\Gamma_{EELS}(\omega) = \frac{e}{\pi\hbar\omega} \int \mathcal{R}\{e^{-i\omega t} \cdot \mathbf{v} \cdot \mathbf{E}^{ind}[\mathbf{r}_e(t), \omega]\} dt. \quad (2)$$

Similarly, the [CL spectrum](#) measures the energy radiated to the far field by the plasmons excited by a moving electron. For the simple geometry of an isolated spherical nanoparticle (see fig. 1(a)), analytical expressions for the loss and [cathodoluminescence](#) spectra induced by a fast electron can be derived (based on Mie scattering theory³⁸), and include relativistic effects:^{25,26}

$$\Gamma_{EELS}(\omega) = \frac{e^2}{c\hbar\omega} \sum_{l=1}^{\infty} \sum_{m=-l}^l K_m^2 \left(\frac{\omega b}{v\gamma} \right) \times [C_{lm}^M \text{Im}(t_l^M) + C_{lm}^E \text{Im}(t_l^E)] \quad (3)$$

$$\Gamma_{CL}(\omega) = \frac{e^2}{c\hbar\omega} \sum_{l=1}^{\infty} \sum_{m=-l}^l K_m^2 \left(\frac{\omega b}{v\gamma} \right) \times [C_{lm}^M |t_l^M|^2 + C_{lm}^E |t_l^E|^2]. \quad (4)$$

where t_l^M and t_l^E are the magnetic and electric Mie theory scattering coefficients, which are independent on the electron velocity and impact parameter. C_{lm}^E and C_{lm}^M are the magnetic and electric coupling coefficients that depend exclusively on the electron velocity (i.e. v/c ratio) and $\gamma = 1/\sqrt{1 - v^2/c^2}$ is the Lorentz factor accounting for relativistic effects and the contraction of impact parameter b as seen by the moving electron (see fig. 1(a)).

For an isolated gold nanosphere with radius $a = 20$ nm (see fig. 1(a)) and impact parameter $b = 22$ nm (i.e. the electron beam is passing 2 nm away from the nanoparticle), Γ_{EELS} and Γ_{CL} are calculated using eq. (3) and eq. (4) and shown in fig. 1(b) and (c) respectively. We immediately observe that Γ_{EELS} and Γ_{CL} have very different dependence on the electron beam velocity v . The normalized Γ_{EELS} decreases with increasing electron beam velocity, while the [cathodoluminescence](#) spectra Γ_{CL} peak at electron velocity $v = 0.2c$ and are red-shifted with response to Γ_{EELS} . These analytical calculations obtained from eq. (3) and eq. (4) are in-line with full-wave finite-difference time-domain numerical simulations as

Figure 1: Normalized loss Γ_{EELS} and radiative Γ_{CL} spectra for (a-c) an isolated metal nanoparticle (NP), (d-f) nanowire on mirror (NWoM) effective 2D nanocavity and (g-i) nanoparticle on mirror (NPoM) 3D nanocavity with (nanowire and nanosphere) radius $a = 20$ nm, gap size $d = 1$ nm and dielectric spacer in the gap of $n = 1.4$. The loss Γ_{EELS} spectra for the three structures are shown as a function of electron beam velocity in (b), (e) and (g) respectively and the radiative Γ_{CL} spectra in (c), (f) and (i).

shown in fig. S1 - Supp. Info.

A systematic analysis of the building blocks of eq. (3) and eq. (4) offers an intuitive understanding on how electrons couple energy into different plasmon modes and how the plasmon modes, in turn, induce the energy loss (Γ_{EELS}) in the beam and emit to the far-field (Γ_{CL}). We identify three main components in eq. (3) and eq. (4): Firstly, the plasmon response to the excitation is mediated by the Mie scattering coefficients $t_l^{M,E}$ which describe how each plasmon mode scatters/absorbs the impinging excitation. The scattering Mie coefficients are independent of the electron velocity, but define the plasmon mode of a spherical nanoparticle (see fig. S2 in Supp. Info for the behavior of each plasmon mode). The second component of eq. (3) and eq. (4) is governed by the coupling coefficients $C_{lm}^{M,E}$, which are a measure of the in-coupling energy to each plasmonic mode induced by the moving electron and are dependent on the electron velocity v . In general, the in-coupling term reduces in value with the electron velocity (fig S3. in Supp. Info). However, for small electron velocities, equivalent energy is coupled into all plasmon modes (bright and dark), while for fast electrons the dipole resonance (i.e. lowest-order mode $l = 1$) acquires more energy due to its bright/radiative nature (see fig. S3 Supp. info). Reciprocity ensures that for bright modes which efficiently radiate energy to the far-field one can also efficiently couple in energy to them. Finally, the third term of eq. (3) and eq. (4) describes the energy that fast electrons inject into the plasmon modes via the modified Bessel function $K_m^2\left(\frac{\omega b}{v\gamma}\right)$ that emits a cylindrical electromagnetic field around the electron trajectory and has a vanishing character away from it. It depends on the electron velocity and, as expected, faster electrons carry more energy (see fig. S4 Supp. Info).

The different dependence of Γ_{EELS} and Γ_{CL} in fig. 1(b) and (c) is therefore due to the in-coupling coefficient $C_{lm}^{M,E}$. At very slow electron velocities ($v \approx 0.1c$), the differences between the [cathodoluminescence](#) spectra in fig. 1(c) and loss spectra in fig. 1(b) are due to the efficient excitation of higher order modes that are dark and dominate the loss spectra Γ_{EELS} , but radiate inefficiently and make an insignificant contribution to Γ_{CL} . This is

one of the main reasons why EELS is widely used to characterize plasmonic devices, because electrons can map high order resonances that are difficult to couple to from the far-field, using for example dark-field microscopy.^{26,39–42} For faster electrons ($v \approx 0.2c$), energy is coupled more efficiently into the radiative lowest order mode ($l = 1$) than higher-order dark plasmons, which leads to a competitive behavior dependent on the electron velocity, nanoparticle size and impact parameter. For even faster and relativistic electrons ($v > 0.5c$), the coupling coefficients $C_{lm}^{M,E}$ gradually reduce in value, which is evident in both the radiative and loss spectra. Hence, while faster electrons carry more energy into the system, they couple less efficiently to all plasmonic modes.

While the description for an isolated metallic nanoparticle provides useful insight about the impact of the electron velocity on the Γ_{EELS} and the Γ_{CL} spectra, most common plasmonic environments are actually coupled nano-structures since they can better confine and enhance electromagnetic fields. Hence, we focus on two types of plasmonic nanocavities, comprising a nanoparticle assembled on a flat metal surface and separated by a molecular monolayer of nanometer thickness: the **nanowire on mirror (NWoM) constituting an effective 2D nanocavity** (fig. 1(d)) and the **nanoparticle on mirror (NPoM)** which is a 3D nanocavity (fig. 1(g)) where strong coupling of a single molecule was realized at room temperature.¹³ For both nanocavities, the gold nanowire/nanoparticle of radius $a = 20$ nm is separated from the gold mirror by a molecular layer with $n = 1.4$,¹ forming nanocavities of 1 nm height. The electron beam propagates parallel to the mirror and never penetrates the metal. We then calculate numerically the loss Γ_{EELS} and the radiative Γ_{CL} spectra for both structures, shown in fig. 1(e)-(f) and (h)-(i). It is immediately evident that the 2D and the 3D nanocavities have very different response to the electron beam velocity. This is due to the different spectral position of the dark plasmon modes for the two structures.

We have recently shown³⁶ that 2D plasmonic nanocavities exhibit dark modes that are spectrally degenerate with the primary bright mode (in this case $\lambda_B = \lambda_D \sim 650$ nm), while 3D nanocavities exhibit a spectrally isolated bright mode (at $\lambda_B \sim 650$ nm) and the dark

modes at shorter wavelengths (at $\lambda_D \sim 550$ nm) – see fig S6 Supp. Info. for more details. For the NPoM, small velocities of $v = 0.05c - 0.15c$ comparably excite all plasmon modes, with Γ_{EELS} showing a main feature at ~ 500 nm where all higher-order dark modes are super-positioned. The loss spectra of the NPoM for this velocity range is in fact comparable to the isolated spherical nanoparticle since most of the higher-order modes are closely confined on the spherical surface. Γ_{CL} shows the bright dipole ($l = 1$) mode at 650 nm, with the second order mode ($l = 2$ - quadrupole) at ~ 580 nm. For increasing electron velocity, more energy is coupled primarily into the radiative ($l = 1$) dipole mode and less into the higher order dark modes, increasing both Γ_{EELS} and Γ_{CL} at 650 nm. The transition electron velocity where the bright modes start to dominate over the dark ones is $v = 0.15c$ for this system, and is determined by the coupling coefficients $C_{lm}^{M,E}$. The same qualitative behavior is observed for the 2D NWoM for velocities $v > 0.2c$, where the radiative dipole mode dominates both Γ_{EELS} and Γ_{CL} at ~ 650 nm. Nevertheless, the spectral degeneracy of the dark and bright plasmon modes produces a stronger and broader resonance. For slow electron velocities ($v < 0.2c$), the dark plasmon mode at ~ 650 nm produces an equivalent loss Γ_{EELS} spectrum to the super-positioned higher-order modes at ~ 550 nm, giving rise to the different spectrum map between the 2D and the 3D nanocavity. This behavior is clearly illustrated in figure S7 in Supp. Info, where the loss spectra at $v = 0.15c$ is dominated by the superposition of higher order modes at ~ 500 nm, especially for the NPoM. Hence, the excitation interplay between the dark and bright plasmons in plasmonic nanocavities significantly impacts the loss and cathodoluminescence spectra observed as a function of the electron velocity, and, as we show below, also changes the Rabi dynamics of QEs with plasmons accordingly.

Results and Discussion

We now place inside the two plasmonic nanocavities a quantum emitter (QE) described by a two-level system with dipole moment $\mu = 3.79$ D, dephasing $\Gamma = 28$ meV and relaxation time

of the excited state $\tau = 1$ ns. The parameters correspond to realistic molecules, for example methylene blue used to realize strong coupling at room temperature in the NPoM configuration^{13,16}. Since the dipole mode ($l = 1$) for the two configurations exist at slightly different wavelengths, we tune the transition wavelength of the QE accordingly to be $\lambda_{12} = 635$ nm for the NWoM and $\lambda_{12} = 650$ nm for the NPoM. The response of the quantized two-level system is described semi-classically using dynamical Maxwell-Bloch equations⁴³ implemented within the classical FDTD calculations, described in more detail in Methods. Thus, the density matrix equations of motion eq. (9) are used to calculate the excited/ground state population (N_2/N_1) at each time step, and the induced macroscopic polarization which is updated self-consistently into the classical FDTD solver, resulting in the classical electromagnetic response due to the presence of the two-level system.

The loss and cathodoluminescence spectra for the 2D NWoM nanocavity with a single QE placed in the center of the nanocavity are shown in fig. 2(a) and (b). As expected, both spectral maps show the characteristic hybrid states and mode splitting. Figure 2(c) and (d) show the loss and the cathodoluminescence spectra for electron velocity of $v = 0.5c$ with and without the presence of the QE, where the characteristic properties of the hybrid states are evident. The electron beam velocity does not significantly change the spectral position of the hybrid states, but impacts the intensity symmetry and the bandwidth of the polaritons. Spectral asymmetries for hybrid states emerge due to the QE dephasing, here summarized within the Γ term of the two-level description which is due to the vibrational states of the QEs.⁴⁴ However, such asymmetries also arise due to the coupling of the QE with dark plasmons,⁴⁴ implying that the electron beam velocity has an impact on the polariton intensity symmetry. The asymmetries of the hybrid states are better observed in fig. 2(e), where the normalized amplitudes of the lower polariton (LP) and upper polariton (UP) are plotted as a function of the electron beam velocity v for both the loss and cathodoluminescence spectra. For the CL spectra, the symmetry of the two polaritons inverts at $v = 0.1c$ and the amplitude increases subsequently with v for $v > 0.4c$, while the loss spectra peaks at

Figure 2: The formed polaritons for a 2D NWoM nanocavity of radius $a = 20$ nm, gap $d = 1$ nm, dielectric medium in the gap with $n = 1.4$ strongly coupled to a QE. The normalized (a) loss Γ_{EELS} and (b) radiative Γ_{CL} spectral maps as a function of the electron beam velocity. For an electron beam velocity of $v = 0.5c$, we plot the (c) loss and (d) cathodoluminescence spectra with a QE emitter present (red) and without (blue). (e) The relative amplitude of the upper (UP) and lower (LP) polaritons extracted from the loss (dotted lines) and cathodoluminescence spectra (solid lines) as a function of the electron beam velocity.

Figure 3: The anti-crossing maps for the [cathodoluminescence spectra](#) showing the strong coupling hybrid states formed by the dipole bright mode (λ_P) of a 2D NWoM nanocavity and a quantum emitter with resonant wavelength λ_{12} , and shown for three electron beam velocities: (a) $v = 0.1c$, (b) $v = 0.5c$ and (c) $v = 0.9c$

$v \sim 0.4c$.

We want to emphasize at this point that, in contrast to a standard high Q-cavity system where only one photonic cavity mode with a narrow bandwidth interacts with the emitter, in a nanoplasmonic system multiple photonic modes with broad bandwidths and dark/bright nature exist, and the quantum emitter is dressed by each of these modes in a different way. The dressing of the QE by each photonic mode independently remains constant (as one expects in a standard photonic system). However each photonic mode is also dressed by the external excitation (electron beam) in a different way. The energy coupled into each photonic mode changes with the electron velocity. Hence, while each photonic mode is still coupled with the same coupling constant to the emitter, the energy channeled into each coupling interaction varies with variation of the electron velocity, causing in turn the overall response of the system to change with the electron velocity.

To uncover the impact of the electron beam velocity on the hybrid states, we now choose representative electron beams with $v = 0.1c$, $v = 0.5c$ and $v = 0.9c$ and calculate the characteristic anti-crossing maps by varying the transition wavelength λ_{12} of the QE. The results are shown in fig. 3 for the [cathodoluminescence spectra](#), where λ_P is the resonant

wavelength of the plasmon, the white dots are the extracted polariton frequencies from the simulations and the solid lines are theoretical hybrid modes calculated using a semi-classical coupled three-oscillators model described in detail in the Methods section. Hence, in order to characterize the hybrid modes of the CL spectra in fig. 3, the fittings of eq. (11) with coupling strengths $g_{v=0.1c} = 60.9 \text{ meV}$, $g_{v=0.5c} = 61.57 \text{ meV}$ and $g_{v=0.9c} = 61.26 \text{ meV}$ were used. The weaker coupling strength at slow velocities is due to the lossy nature of the dark mode that are more favorably excited for small v . We remark that the anti-crossing and coupling strength remains relatively robust for a broad range of electron beam velocities.

The near-field dynamics of the same system plotted in fig. 4(a) and (c) are more revealing, showing the characteristic cyclic energy exchange observed in the strong coupling regime. To isolate the fields due to the emitter-plasmon interaction alone, we subtract the E-field at the QE position when the two-level is present ($E^{em}(t)$) and not present ($E^{cav}(t)$): $E^{ind}(t) = E^{em}(t) - E^{cav}(t)$, and plot the resulting oscillations in fig. 4(a).

The induced dynamics changes considerably with the electron beam velocity, with significant differences in the waveform and the period of the Rabi oscillations. The Rabi frequencies obtained from the near field (E^{ind}), the radiative (Γ_{CL}) and the loss (Γ_{EELS}) spectra are summarized in fig. 4(c). The near-field Rabi-frequencies are extracted from the excursion envelope of E^{ind} and taking the mean of the oscillation period of each cycle as $\tau_\mu = \frac{1}{N} \sum_{i=1}^N \tau_i$, where N is the number of Rabi cycles, while the spectral frequencies are extracted from fig. 2(a) and (b). At slow electron beam velocities, there is a difference of $\sim 70 \text{ meV}$ for the Rabi frequency extracted from the near-field and the cathodoluminescence spectra in the far-field. The near-field spectra also contain Rabi oscillations between the QE and the dark plasmon modes, which, as we reported earlier,³⁶ have a smaller Rabi frequency, However, this process is never transmitted to the far-field and therefore the radiative spectra cannot record it. As the electron beam velocity increases, the dark plasmons are less efficiently excited, and thus the near-field and the radiative Rabi frequencies converge. In fact, they reach similar values to the Rabi frequencies recorded by the loss spectra, which records

Figure 4: Near-field strong coupling dynamics for (a) the 2D NWoM and (b) the 3D NPoM nanocavities. The Rabi frequency (Ω) extracted from the near-field dynamics (blue curve), the loss (green curve) and radiative (red) spectra are shown as a function of electron beam velocity v for (c) the 2D NWoM and (d) the 3D NPoM nanocavities.

both dark and bright plasmons. Hence, the strong coupling dynamics are in fact sensitive to the excitation of the system, even if spectrally appear to be robust, with more energetic electrons inducing faster Rabi oscillations.

To confirm the conclusions above, we perform the same investigation for the NPoM nanocavity, which exhibits a spectrally isolated bright mode at $\lambda = 650$ nm (see Supp. Info. fig. S6). The near-field strong coupling dynamics are shown in fig. 4(b) as a function of the electron beam velocity. Compared with the 2D NWoM nanocavity, the Rabi oscillations have a longer period, and are mostly independent of the electron beam velocity. Figure 4(d) summarizes the Rabi frequencies obtained at the near-field, the radiative and the loss spectra, and all show similar values and hence are independent of the electron beam velocity. The smaller Rabi frequency value for the 3D NPoM is due to the larger mode volume for 3D plasmonic structures that confines the electromagnetic fields less.

To sum up the results in fig. 4, the complex time dynamics of the QE is captured differently in the EELS and CL spectra. Thus, for the NWoM, the loss spectra reflects the interaction of the emitter with the spectrally degenerate bright and dark dipole mode, while the CL spectra captures only the interaction with the bright one. In the case of the NPoM where the emitter is resonant only with the bright plasmon mode, the Rabi frequency extracted from both loss and cathodoluminescence spectra is consistent with the quantum emitter dynamics.

Finally, the full radiative and loss maps are plotted in in fig. 5(a) and (b), where the polaritons are shown as a function of the electron beam velocity for the 3D NPoM nanocavity. Unlike the 2D nanocavity, for the 3D nanocavity the QE exchanges energy with only one mode, the bright dipole ($l = 1$) plasmon. Therefore, the asymmetry of the polaritons never inverts and, in fact, remains robust with increasing the electron velocity (fig. 5(e)) due to the absence of the dark mode.

In conclusion, we have demonstrated that one can dynamically control, and even modify, the ultrafast Rabi oscillation dynamics between a QE and a plasmonic nanocavity via the

Figure 5: The formed polaritons for a 3D NPoM nanocavity of radius $a = 20$ nm, gap $d = 1$ nm, dielectric medium in the gap with $n = 1.4$ strongly coupled to a QE. The normalized (a) loss Γ_{EELS} and (b) radiative Γ_{CL} spectral maps as a function of the electron beam velocity. For an electron beam velocity of $v = 0.5c$, we plot the (c) loss and (d) [cathodoluminescence spectra](#) with a QE emitter present (red) and without (blue). (e) The relative amplitude of the upper (UP) and lower (LP) polaritons extracted from the loss (dotted lines) and [cathodoluminescence spectra](#) (solid lines) as a function of the electron beam velocity.

velocity of a passing electron beam, given a dark plasmon is present. We initially decompose the analytical description for a spherical nanoparticle excited by a moving electron beam to understand how different plasmonic modes are excited, absorb energy and radiate to the far-field. We find that at slow electron velocities, there is a competitive behavior for the in-coupling energy into the dark and bright plasmons, which results in different radiative and loss spectra for different electron beam velocities. Plasmonic nanocavities that confine light in extremely small volumes support a complex set of dark and bright modes, which can be used to dynamically modify and control the Rabi oscillations. We show that when dark modes are present, the Rabi oscillations in the near-field can be twice slower than the Rabi frequency observed in the loss and [cathodoluminescence spectra](#) commonly measured experimentally. This discrepancy is dominant for the low electron velocities, where dark modes are efficiently excited and dominate the near-field dynamics, and all frequencies converge as the electron beam velocity increases. This behavior is not present when only bright modes are involved in the interaction with the QE. In this way, we extend the coherence time available for strong coupling by exploring the long-lived quantum coherences characteristic of the dark plasmon modes. Hence, we show that using an electron beam excitation one can dynamically control the Rabi frequency by tuning the electron beam velocity, thus paving a dynamical route for strong coupling in plasmonics.

Methods

Maxwell-Bloch Theory

The resonant light-matter interaction of the quantum emitter with the plasmonic system is described via the dynamical Maxwell-Bloch equations.⁴³ These are a semi-classical model which describe the interaction between a multi-level system and a classical light-field via the density matrix equations of motion. Another essential simplification is the dipole approximation; since the electrons in the atom or molecule are confined on a length scale much smaller

than the wavelength of light which is resonant to the transition energy, this approximation is almost always applicable to describe atomic or molecular transitions.

The starting point for the derivation of the Bloch equations is a semi-classical Hamiltonian including an atomic Hamiltonian $\hat{H}_0$ and a time-dependent interaction of the atom with the electromagnetic field $\hat{H}_I$ described within the dipole approximation⁴³

$$\begin{aligned}\hat{H} &= \hat{H}_0 + \hat{H}_I \\ &= \hbar\Omega\hat{\sigma}^\dagger\hat{\sigma} - \hat{\mu} \cdot \mathbf{E}\end{aligned}\tag{5}$$

where $\Omega = (E_2 - E_1)/\hbar$ is the transition frequency between the two electronic states, $\hat{\mu} = \langle 1|\mathbf{r}|2\rangle$ the dipole matrix element of the electronic transition and $\hat{\sigma} = |1\rangle\langle 2|$ is the exciton annihilation operator, $\mathbf{E}$ is the electric field vector calculated classically from Maxwell equations. The semi-classical evolution of a multi-level electronic system driven by a classical field is obtained from the von Neumann equation:

$$\frac{d\hat{\rho}}{dt} = -\frac{i}{\hbar} [\hat{H}, \hat{\rho}] + \frac{\gamma_r}{2} (2\hat{\sigma}\hat{\rho}\hat{\sigma}^\dagger - \hat{\sigma}^\dagger\hat{\sigma}\hat{\rho} - \hat{\rho}\hat{\sigma}^\dagger\hat{\sigma}) + \frac{\gamma_p}{2} (2\hat{\sigma}^\dagger\hat{\rho}\hat{\sigma} - \hat{\sigma}\hat{\sigma}^\dagger\hat{\rho} - \hat{\rho}\hat{\sigma}\hat{\sigma}^\dagger) + \frac{\gamma_d}{2} (\hat{\sigma}_z\hat{\rho}\hat{\sigma}_z - \hat{\rho}),\tag{6}$$

where $\hat{\rho}$ is the density matrix and γ_r , γ_p and γ_d denote the incoherent relaxation, pumping and pure dephasing rates respectively.⁴³ Solving for the Hamiltonian in eq. (5), the density matrix equation can be reduced to:⁴⁵

$$\begin{aligned}\frac{\partial\rho_{12}}{\partial t} &= -(\Gamma - i\omega_{12})\rho_{12} - i\frac{\mu \cdot \mathbf{E}}{\hbar}(\rho_{22} - \rho_{11}) \\ \frac{\partial\rho_{22}}{\partial t} &= -\frac{\partial\rho_{11}}{\partial t} = -\gamma\rho_{22} - 2\frac{\mu \cdot \mathbf{E}}{\hbar}\mathcal{I}(\rho_{12}),\end{aligned}\tag{7}$$

where $\Gamma = \gamma_d + \frac{\gamma_r}{2} + \frac{\gamma_p}{2}$ and $\gamma = \gamma_p + \gamma_r$. Using the definition for the induced macroscopic polarization $\mathbf{P} = N_d \text{Tr}(\rho_{12}\mu) = 2N_d\mu\mathcal{R}(\rho_{12})$ with N_d the total carrier density per unit volume and defining the excited- (N_2) and ground-state (N_1) populations as the occupations of the density matrix ρ_{22} and ρ_{11} , respectively, the dynamical Maxwell-Bloch equations

reduce to

$$\frac{\partial^2 \mathbf{P}}{\partial t^2} + 2\Gamma \frac{\partial \mathbf{P}}{\partial t} + (\Gamma^2 + \omega_{12}^2) \mathbf{P} = -\frac{2\omega_{12}|\mu|^2}{\hbar} (N_2 - N_1) \mathbf{E} \quad (8)$$

$$\frac{\partial N_2}{\partial t} = -\frac{\partial N_1}{\partial t} = -\gamma N_2 + \frac{1}{\hbar\omega_{12}} \left(\frac{\partial \mathbf{P}}{\partial t} + \Gamma \mathbf{P} \right) \cdot \mathbf{E}. \quad (9)$$

The induced polarization $\mathbf{P}$ is driven by the local field $\mathbf{E}$ at the position of the QE and is updated self-consistently in the FDTD Maxwell's equations solver, describing the coupling back to the plasmon modes. For the results presented in this paper, we set $\mu_x = 3.79 \text{ D}$, $\Gamma = 28 \text{ meV}$ and $\gamma = 10^9 \text{ Hz}$, corresponding to a relaxation time of the excited state $\tau = 1 \text{ ns}$.

Strong Coupling in a Semi-Classical Description

Various phenomenological models have been proposed to quantitatively explain the strong coupling between the plasmon modes and the two-level emitter. The simplest one describes the coupling between the plasmon mode and the quantum emitter as a classical system of coupled Lorentz oscillators⁴⁶ and the resulting hybrid modes are derived from the harmonic motion of the coupled system. Nevertheless, this description implies that the emitter couples exclusively to one plasmon mode, while it is clear from the spectra in fig. 2 that the higher order modes, especially the quadrupole, are also playing an important role. One can describe the interaction between the emitter exciton mode and the two plasmonic modes using a phenomenological three coupled oscillators model. The uncoupled plasmon and emitter resonances are given by a diagonal matrix H_{nc} , while the coupling between each plasmon mode and the emitter is given via the off-diagonal matrix H_c .⁴⁷ Putting everything together, the total system can be described by

$$H = H_{nc} + H_c = \begin{pmatrix} \Omega_{pl_1} & 0 & g_1 \\ 0 & \Omega_{pl_2} & g_2 \\ g_1 & g_2 & \Omega_{QE} \end{pmatrix}. \quad (10)$$

$\Omega_{QE} = \omega_{12} - i\Gamma$ and $\Omega_{pl_x} = \omega_x - i\frac{\omega_x}{2Q_x}$ are the complex frequencies of the emitter and plasmon mode x . Γ is the emitter linewidth, while Q_x is the quality factor of the plasmon cavity mode x . g_1 and g_2 are the coupling strengths between the emitter and the first and second plasmon mode, respectively. We also assume that there is no coupling between the two plasmon modes. The three hybrid modes are given by the solutions to the characteristic equation:

$$\det(H - \mathbb{1}\omega) = 0 \quad (11)$$

The coupling strength between a quantum emitter within the dipole approximation and a resonator mode is

$$g = \mu \sqrt{\frac{\pi c \hbar}{\lambda_{12} \epsilon_0 n^2}} \sqrt{\frac{N}{V}}, \quad (12)$$

where $\mu = 3.8 \text{ D}$ is the dipole moment of the emitter, λ_{12} is the resonant wavelength of the QE, $n = 1.4$ is the refractive index of the cavity medium, N is the number of emitters and V is the mode volume of the respective electromagnetic mode. The real part of the emitter frequency ω_{12} is varied between 500 and 850 nm, which gives the range for $\lambda_{12} = 2\pi c/\omega_{12}$; $\Gamma = 28 \text{ meV} \approx 6.77 \text{ THz}$ is the linewidth of the two-level transition.

Using all the above parameters, one can find the eigenvalues of the coupling matrix as a function of the emitter wavelength which give the hybrid modes between the emitter and the plasmon modes. The only fitting parameters are the mode volume V and the quality factor Q . The values used are $Q_1 = 20$ and $V_1 = 15 \text{ nm}^3$ for the dipole mode and $Q_2 = 10$ and $V_2 = 20 \text{ nm}^3$ for the quadrupole mode.

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Supporting Information Available

The Supporting Information file is available free of charge on ... at DOI:

- Filename: The loss and [cathodoluminescence spectra](#) for an isolated nanoparticle calculated using Mie theory (analytically) and numerically; step-by-step description of how electrons couple energy into different plasmon modes; the nanoparticle radius impact on the loss and [cathodoluminescence spectra](#); The bright and dark plasmon modes absorption cross-sections for both the 2D NWoM and 3D NPoM nanocavities.

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