Controlling Single-Photon Emission with Ultrathin Transdimensional Plasmonic Films

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The properties of a two-level quantum dipole emitter near an ultrathin transdimensional plasmonic film are studied theoretically. The model system studied mimics a solid-state single-photon source device. Using realistic experimental parameters, the spontaneous and stimulated emission intensity profiles are computed as functions of the excitation frequency and film thickness, followed by the analysis of the second-order photon correlations to explore the photon antibunching effect. It is shown that ultrathin transdimensional plasmonic films can greatly improve photon antibunching with thickness reduction, which allows one to control the quantum properties of light and make them more pronounced. Knowledge of these features is advantageous for solid-state single-photon source device engineering and overall for the development of the new integrated quantum photonics material platform based on the transdimensional plasmonic films.

1. Introduction

Transdimensional (TD) quantum materials are atomically-thin films of precisely controlled thickness, films made of precisely controlled finite number of monolayers,[13–3] The term “trans-dimensional” refers to the transitional range of thickness—a regime that is neither three (3D) nor two (2D) dimensional but rather something in between, turning into 2D as thickness tends to zero, challenging to study what the 3D-to-2D continuous transition has to offer to improve material functionalities. Currently available due to the rapid progress in nanofabrication techniques,[4–11] such materials offer high tailorbility of their electronic and optical properties not only by altering their chemical and/or electronic composition (stoichiometry, doping) but also by merely varying their thickness (number of monolayers).[12–15] Materials like these are indispensable for studies of fundamental properties of the light–matter interaction as it evolves from a single 2D atomic layer to a larger number of layers approaching the 3D bulk material properties. With thickness of only a few atomic layers, ultrathin TD films of metals, doped semiconductors, or polar materials can support plasmon-, exciton-, magnon-, and phonon-polariton eigenmodes.\[14–32\] Plasmonic TD materials (ultrathin metallic films) offer controlled light confinement, large tailorbility and dynamic tunability of their optical properties due to their thickness-dependent localized surface plasmon (SP) modes\[14–21\] which are distinctly different from those of conventional thin films commonly described by either purely 2D or by 3D material properties with boundary conditions imposed on their top and bottom interfaces.\[13–42\]

In such systems, the vertical quantum confinement enables a variety of new quantum phenomena, including the thickness-controlled plasma frequency red shift\[2,11\] the SP mode degeneracy lifting\[14,18\] a series of magneto-optical effects\[13\] and even atomic transitions that are normally forbidden\[1,20,21\] to mention a few.

Previously, we have implemented the confinement-induced nonlocal Drude response model based on the Keldysh–Rytova (KR) pairwise electron interaction potential\[22\] to study theoretically the electronic properties of ultrathin TD plasmonic films\[13,14\] and demonstrate their major manifestations experimentally.\[11,42\] The KR interaction potential takes into account the vertical electron confinement\[13,44\] which makes it much stronger than the electron–electron Coulomb potential\[43\] offering also the film thickness as a parameter to control the nonlocal optical response of the TD plasmonic films. The optical response nonlocality was previously shown experimentally to be a remarkable feature of quantum-confined plasmonic nanostructures\[12,45\] Here, we use our nonlocal KR model to further explore the capabilities of the TD plasmonic films, focusing on the light-scattering properties of a quantum dipole emitter (DE) placed at a distance near the film surface; see the inset in Figure 1b. The model system we study mimics a quasi-2D solid-state single-photon source device\[46\] for which our goal is to show the advantages of using the ultrathin TD plasmonic films. So far room-temperature solid-state quantum emitters have been observed in wide-bandgap semiconductors such as diamond\[47\] and silicon carbide,\[48\] nanocrystalline quantum dots,\[49–51\] and in carbon nanotubes\[52\] Single-photon emission from localized color-center defects in 2D materials has been reported both at cryogenic\[53–56\] and at room
temperatures.[57,58] An aspiring goal, however, is to use these materials in the quantum regime where the photon emission is of robust and controllable single-photon character, to enable applications in quantum information processing.[46,59] We show that the ultrathin TD plasmonic films have this outstanding potential due to their unique tailoring by means of their thickness adjustment.

Using the nanodiamond nitrogen-vacancy (NV) center[60,61] near the TiN film surface as a prototype coupled “DE-TD film” system,[14] we compute the spontaneous and stimulated emission intensity profiles as functions of the excitation frequency and film thickness, followed by the intensity correlation function analysis to explore the photon antibunching effect. Transition metal nitrides have emerged as a new class of materials with great promise to substitute noble metals such as gold and silver,[62] which have exceptional plasmonic properties but relatively low melting temperatures making them incompatible with semiconductor fabrication technologies. Titanium nitride, in particular, has low-loss plasmonic response and high melting point, on the contrary, making it structurally stable TD material capable of forming stochiometrically perfect ultrathin films of controlled thickness down to 1 nm at room temperature.[11] Here we show that the film thickness can be used to tune from weak to strong the evanescent coupling of the DE to the plasma modes of the ultrathin TD films. The strong evanescent coupling hybridizes the transition energy levels of the DE whereby its stimulated emission is quenched. Increased resonance optical pumping reduces this quenching effect. The overall film thickness effect on the intensity correlation function is to increase its positive slope with thickness reduction and thus to improve the photon antibunching and related (nonclassical) sub-Poissonian photon counting statistics.[63] Knowledge of these features is advantageous for solid-state single-photon source device engineering[46] and overall for the development of the new integrated quantum photonic material platform[64] based on the ultrathin TD materials for quantum information processing applications.

2. Nonlocal Dielectric Response and Spontaneous Emission Spectrum

If the environment has a lower dielectric constant than that of the film, such as the case of the finite-thickness planar metallic structure shown in the inset of Figure 1b with \( \varepsilon_1,2 < \varepsilon \), then the increased “outside” contribution makes the Coulomb interaction of electrons vertically confined stronger than that in a homogeneous medium with the same dielectric constant.[43] As a consequence, the in-plane plasma oscillation frequency of the film takes the form[42]

\[
\omega_p = \frac{\omega_p^{3D}}{\sqrt{1 + (\varepsilon_1 + \varepsilon_2)/k d}},
\]

where \( k \) is the in-plane momentum absolute value and \( d \) is the film thickness. This makes the Drude dielectric response function of the plasmonic film

\[
\frac{\alpha(k, \omega)}{\varepsilon} = 1 - \frac{\omega_p^{3D}}{\alpha(\omega + i \gamma)}
\]

\( k \)-dependent, or nonlocal. Here, \( \gamma \) is the phenomenological inelastic electron scattering rate. As \( d \) decreases, \( \omega_p(k) \) shifts to the red and Equation (2) acquires the \( 1/k \) nonlocal spatial dispersion of 2D materials. In the TD regime, for ultrathin plasmonic films of finite thickness, the low-frequency response is controlled by Equations (1) and (2), with \( \varepsilon \approx 10 \) contributed by both positive ion background and interband transitions.[11,42] As
Rescaling the equations above with substitutions representing the evanescent and propagating wave contributions dominant near the surface and at large distances from the surface of the film, respectively.

Figure 1a shows the earlier reported plasma mode distribution derived from the p-evanescent wave reflection coefficient of the ultrathin TD plasmonic film as a function of the thickness \( d \) of the film.\(^{14}\) The associated spontaneous emission rate is shown in Figure 1b as a function of \( d \) and the DE position \( z_A \) near the film surface where the evanescent contribution to the spontaneous emission is dominant. This graph is obtained from Equation (3) with Green tensor Equation (4) approximated by the first term of Equation (7) averaged over the three dipole orientations and normalized by the free-space spontaneous emission rate \( \Gamma_0 \). The DE transition frequency \( \omega_A \) is taken to be 1.81 eV consistent with what was reported recently from the density functional theory simulations for the zero-phonon line transition of the NV-center in diamond,\(^{61}\) with all other parameters being essentially representative of the ultrathin TiN film as reported earlier.\(^{14}\) Comparing the (a) and (b) panels in Figure 1, it can be seen that the fast increase followed by the drop-off of the DE spontaneous emission rate at \( d < 10 \text{ nm} \) originates from the DE coupling to the split-off plasma mode of the lowest out-of-plane momentum which is present in there for \( 10 \geq d \geq 5 \text{ nm} \). The presence itself of this split-off mode is a remarkable feature of the confinement-induced nonlocality of the KR model we use.\(^{11,14}\)

To see how the DE coupling to the plasma modes in Figure 1a affects its spontaneous emission intensity profile, we use the analytical approach developed previously for an analogous problem of the DE coupled to a resonance of the local density of photonic states (LDOS) close to the nanotube surface.\(^{67}\) As can be seen in Figure 1a, we now have a single plasma resonance for each \( d \) value fixed. In our notations here, the spectral lineshape profile of the spontaneous emission intensity is given by

\[
I(d, z_A, \omega) = \frac{\Gamma(d, z_A)}{2\pi} \frac{(\omega - \omega_A)^2 + \delta\omega^2}{\left((\omega - \omega_A)^2 - X^2/4\right)^2 + \delta\omega^2(\omega - \omega_A)^2},
\]

\[
X(d, z_A) = \sqrt{2\delta\omega_0\Gamma(d, z_A)}
\]

Here, \( \Gamma(d, z_A) = \Gamma(d, z_A, \omega_A)/\Gamma_0 \) is the dimensionless spontaneous emission rate presented in Figure 1b and all frequency quantities are assumed to be dimensionless (divided by \( \Gamma_0 \)) including the half-width-at-half maximum \( \delta\omega_0 \) of the LDOS resonance. The parameter \( X \) is the Rabi-splitting to represent the DE transition level hybridization due to the coupling to the medium-assisted excitations of the material subsystem (plasma modes of the TD film herein). The coupling is termed weak if \( (X/\delta\omega)^2 \ll 1 \) and strong if \( (X/\delta\omega)^2 \gg 1 \), in which cases Equation (8) generates the single-peaked \( \omega_A \)-centered Lorentzian profile and the double-peaked profile of two symmetrically split peaks at \( \omega_A \pm X/2 \), respectively.\(^{67}\)

### 3. Stimulated Emission Spectrum

In resonance-fluorescence experiments, the DE is irradiated by laser light tuned to the DE transition frequency, so that the DE can be excited into an upper quantum state. The excitation is followed by competing processes. They are the (coherent)

\[
\Gamma(d, z_A, \omega_A) = \frac{\Gamma(d, z_A)}{2\pi} \frac{(\omega - \omega_A)^2 + \delta\omega^2}{\left((\omega - \omega_A)^2 - X^2/4\right)^2 + \delta\omega^2(\omega - \omega_A)^2},
\]

\[
X(d, z_A) = \sqrt{2\delta\omega_0\Gamma(d, z_A)}
\]
driving of the DE transition by the laser field, thereby stimulated (coherent) photon emission and absorption of a laser photon take turns, and spontaneous (incoherent) emission of the DE excited state. An interplay of the two, of which one is strongly dependent in our case (Figure 1), is expected to give rise to new quantum-statistical features of the scattered radiation.

It was first recognized by Feynmann et al.\(^{[68]}\) that the resonance-fluorescence process can be described by optical Bloch equations, a generalization of the Bloch equations of the magnetic resonance,\(^{[69]}\) whereby the DE stimulated emission spectrum under resonance excitation can be shown to take the form as follows\(^{[65]}\)

\[
S(\omega, \Gamma_{1}) = \frac{1}{\pi} \text{Re} \left\{ \hat{S}_{12} \left[ i \left( \omega - \omega_{1} \right) + \frac{\Gamma_{1}}{2} \right] \right\}
\]

where \(\omega_{1}\) is the laser field frequency, \(\Gamma_{1}\) is the spectral apparatus passband width, and

\[
\hat{S}_{12}(t) = \frac{\sigma_{22}(\infty)}{s + \Gamma_{2}} \left\{ 1 + \frac{\Gamma_{1}}{2s} \left[ 1 - \frac{\Omega/\Gamma_{1}}{(s + \Gamma_{1}) + (s + \Gamma_{2})/\Omega} \right] \right\}
\]

is the Laplace transform of the spectral distribution function under the resonance (\(\omega_{0} = \omega_{1}\) single-photon transition \(1\)→\(2\))\((n + 1)\)\(→\)(\(n + 2\))\(→\)\(n\) \((n\) is the photon occupation number) between the lower \(1\) and upper \(2\) DE states. In this equation, \(\Omega = |d_{z1} \cdot \mathbf{E}_{z1}|/2\hbar\) is the Rabi frequency to represent the DE transition dipole coupling strength to an external laser field, \(\Gamma_{1}\) and \(\Gamma_{2}\) are the longitudinal and transverse relaxation rates, respectively, analogous to those of the Bloch magnetic resonance theory.\(^{[69]}\) In our case here, we have \(\Gamma_{1} = 2\Gamma_{2} = \Gamma(d, z_{1})\) since major relaxation comes due to spontaneous emission from the upper DE state.\(^{[65]}\) The prefactor

\[
\sigma_{22}(\infty) = \frac{1}{2} \frac{\Omega_{1}^{2}}{\Gamma_{1} \Gamma_{2} + \Omega_{1}^{2}}
\]

is the time-dependent upper-state occupation probability taken at \(t = \infty\). All quantities in Equations (10)–(12) are assumed to be dimensionless as per convention introduced above for Equation (8).

Equations (10)–(12) allow for two distinct limiting cases.\(^{[65]}\) They are: 1) the weak-driving-field limit, where \(\Omega \ll \Gamma_{1}, \Gamma_{2}\) to result in the \(\omega_{0}\)-centered \((\omega_{0} = \omega_{1})\) single-peak Rayleigh scattering spectral profile; and 2) the high-driving-field limit, where \(\Omega \gg \Gamma_{1}, \Gamma_{2}\) and the spectrum is reconstructed due to the “dressed” state formation to yield the Mollow triplet profile consisting of the \(\omega_{0}\)-centered main peak and the two symmetric satellites at \(\pm \omega_{0}\).

4. Intensity Correlation and Photon Antibunching

The photon antibunching effect in resonance fluorescence from a quantum two-level DE comes from the fact that having emitted a photon to be detected, the DE undergoes a quantum jump from the upper to the lower (ground) quantum state. The DE cannot emit a second photon from the ground state. Only after performing a laser-induced transition back to the upper quantum state, after absorbing a laser-beam photon, the DE is ready to emit a second photon to be detected. This quantum light emission where the two-level DE produces single photons sequentially one after another is the essence of the photon antibunching, the effect which is the direct opposite of photon emission by bunches that classical light sources do. The probability of emitting the first photon at an instant of time \(t\) is proportional to the upper-state photon occupation probability \(\sigma_{22}(t)\), which also controls the intensity \(S\) of the scattered light as per Equations (10)–(12), and which can be found from the optical Bloch equations to have the form as follows\(^{[65]}\)

\[
\sigma_{22}(t) = \sigma_{22}(\infty) \left( 1 + \frac{\lambda_{1e}^{\dagger}e^{i\omega t} + \lambda_{2e}^{\dagger}e^{i\omega t}}{\lambda_{1} - \lambda_{2}} \right)
\]

Here

\[
\lambda_{1,2} = -\frac{1}{2} (\Gamma_{1} + \Gamma_{2}) \pm \sqrt{\frac{1}{4} (\Gamma_{1} - \Gamma_{2})^{2} - \Omega^{2}}
\]

and the DE is assumed to be resonantly \((\omega_{0} = \omega_{1})\) excited out of its initial lower (ground) quantum state. Similarly, the probability of emitting a second photon at a later instant of time \(t + \tau\) is proportional to the DE upper-state occupation probability \(\sigma_{22}(t + \tau)\) under the condition that at time \(t\) the DE is in its lower (ground) quantum state. Hence, the probability of emitting a first photon at time \(t\) and a second photon at time \(t + \tau\) is proportional to \(\sigma_{22}(t)\sigma_{22}(t + \tau)\). Under the steady-state observation conditions, this turns into the two-photon scattered-intensity correlation function \(\sigma_{22}(\infty)\sigma_{22}(t + \tau)\), a quantity directly related to the normalized second-order autocorrelation function

\[
g^{(2)}(\tau) = \frac{\sigma_{22}(t + \tau)}{\sigma_{22}(\infty)} = 1 + \frac{\lambda_{2e}^{\dagger}e^{i\omega t} + \lambda_{1e}^{\dagger}e^{i\omega t}}{\lambda_{2} - \lambda_{1}}
\]

with \(a = \lambda_{1}/(\lambda_{2} - \lambda_{1})\), commonly used in single-photon source characterization experiments.\(^{[57]}\)

The function \(g^{(2)}(\tau)\) can be seen to have a positive initial slope, which reflects the quantum optics effect of photon antibunching. Clearly, the greater this slope is, the more pronounced quantum features of light are. In addition, it can be shown that since \(g^{(2)}(\tau) < 1\), the scattered fluorescence light gives rise to a (non-classical) sub-Poissonian photocounting statistics.\(^{[63]}\) Both photon antibunching nature and the sub-Poissonian statistics of the resonance fluorescence from a two-level DE system may be regarded as proofs of the quantum nature of light which cannot be understood and explained in terms of classical optics.

5. Discussion of Numerical Results

Figure 2a presents the spontaneous emission spectrum calculated from Equation (8) with \(\delta \omega_{0} = 5\) (taken as a model parameter) and \(\Gamma(d, z_{1})\) shown in Figure 1b for the DE positioned at \(z_{1} = 20\) nm and coupled to the TD film plasma modes shown in Figure 1a. Figure 2b presents the same for \(\delta \omega_{0} = 10\) to compare. The most interesting feature there is that as \(d\) decreases, in the domain \(20 \geq d \geq 5\) nm, the DE-plasmon coupling strengthens to result in the fast spontaneous emission rate increase, as can be seen in Figure 1b, whereby the coupled system is driven in the strong coupling regime \((X/\delta \omega_{1})^{2} \gg 1\) where the new
followed by making it transparent in the reduce this domain in Figure 1a, which dies with dimension profiles in Figure 2 as only one plasma mode is found in sharp central peaks for both spontaneous and stimulated emission profiles into the double-picked profile of the two symmetrically split resonances at \( \omega_A \pm X/2 \). This turns the single-peaked \( \omega_A \)-centered Lorentzian spontaneous emission profile into the double-picked profile of the two symmetrically split resonances at \( \omega_A \pm X/2 \). Increased \( \delta \omega_1 = 10 \) does not eliminate but just smooths out this effect. In this domain of film thicknesses, \( \omega_A \) is no longer the DE eigen transition frequency, and so the intensity of the stimulated emission Rayleigh (same frequency) scattering profile drops down, as can be seen in Figure 2c for the same \( z_A \) and \( \Omega = 1 \), the DE radiative coupling to the external laser field. Instead, it can be shown that the plasmon enhanced Raman (shifted frequency) scattering is very efficient in this regime.\(^\text{[90]}\) Figure 2d shows that the increased DE-laser field coupling, \( \Omega = 10 \), can compete with the DE-plasmon coupling to restore the stimulated Rayleigh scattering intensity by driving the system in the strong radiative coupling regime with “dressed” states formed where the representative Mollow triplet spectral profile appears. Figure 2e, where \( z_A \) is decreased down to 10 nm, demonstrates that bringing the DE closer to the film surface does drop down the intensity of the stimulated emission Rayleigh scattering profile for \( \Omega = 1 \). The obvious reason is that the DE-plasmon coupling increases as well (not shown) so that even increased \( \Omega = 10 \) can barely restore it completely as can be seen from Figure 2f. Finally, decreasing \( d \) below 5 nm restores the sharp central peaks for both spontaneous and stimulated emission profiles in Figure 2 as only one plasma mode is found in this domain in Figure 1a, which dies with \( d \) decreasing to reduce \( \Gamma(d, z_A) \) as can be seen in Figure 1b. This brings our system back in the weak DE-plasmon coupling regime, \( (X/\delta \omega_1)^2 \ll 1 \), followed by making it transparent in the \( d \to 0 \) limit.

Figure 2 presents our calculations of the normalized second-order autocorrelation function \( g^{(2)}(\tau) \) of Equation (15) for two representative \( \Omega = 1 \) and 10 with \( \Gamma(d, z_A) \) shown in Figure 1b, for the DE placed at \( z_A = 10 \) and 20 nm and coupled to the TD film plasma modes shown in Figure 1a. In all four graphs, the left panels show \( g^{(2)}(\tau) \) as functions of \( d \) and \( d, z_A \) shown in Figure 1b for the DE position at \( z_A = 20 \) nm and coupled to the TD film plasma modes shown in Figure 1a. e,f) Same as in (c,d) for the DE position at \( z_A = 10 \) nm.

The obvious reason is that \( \omega_A \) is no longer the DE eigen transition frequency, and so the intensity of the stimulated emission Rayleigh (same frequency) scattering profile drops down, as can be seen in Figure 2f. Finally, decreasing \( d \) below 5 nm restores the sharp central peaks for both spontaneous and stimulated emission profiles in Figure 2 as only one plasma mode is found in this domain in Figure 1a, which dies with \( d \) decreasing to reduce \( \Gamma(d, z_A) \) as can be seen in Figure 1b. This brings our system back in the weak DE-plasmon coupling regime, \( (X/\delta \omega_1)^2 \ll 1 \), followed by making it transparent in the \( d \to 0 \) limit.

Figure 3 presents our calculations of the normalized second-order autocorrelation function \( g^{(2)}(\tau) \) of Equation (15) for two representative \( \Omega = 1 \) and 10 with \( \Gamma(d, z_A) \) shown in Figure 1b, for the DE placed at \( z_A = 10 \) and 20 nm and coupled to the TD film plasma modes shown in Figure 1a. In all four graphs, the left panels show \( g^{(2)}(\tau) \) as functions of \( d \) and \( d, z_A \) shown in Figure 1b for the DE position at \( z_A = 10 \) nm.

6. Conclusion

In this work we use the parameters of the nanodiamond NV center near the TiN film surface to compute the spontaneous...
and stimulated emission intensity profiles as functions of the excitation frequency and film thickness, followed by the intensity correlation function analysis to explore the photon antibunching effect for the quantum DE coupled to the ultrathin TD plasmonic film. We show that the film thickness can be used to tune the evanescent coupling of the DE to the plasma modes of the film. The strong evanescent coupling hybridizes the transition energy levels of the DE, whereby its stimulated emission Rayleigh scattering profile is quenched. Increased resonance optical pumping drives the system in the strong radiative coupling regime, which competes with the DE-plasmon coupling, to restore the stimulated Rayleigh scattering intensity. The overall film thickness effect on the second-order photon autocorrelation function is to increase its positive slope with thickness reduction and thus to improve the photon antibunching properties and related (nonclassical) sub-Poissonian photon counting statistics. Our theoretical predictions can be tested in experiments similar to those reported recently for epitaxial TiN films with thicknesses below 10 nm grown on MgO substrates and covered with an AlScN passivation layer,[1] with nanodiamond NV-centers as DEs which can be deposited right on the varied-thickness passivation layer. Knowledge of the features we predict is advantageous for solid-state single-photon source device engineering and overall for the development of the new integrated quantum photonics material platform based on the ultrathin TD materials for quantum information processing applications.

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Conflict of Interest
The author declares no conflict of interest.

Data Availability Statement
The data that support the findings of this study are available from the corresponding author upon reasonable request.

Keywords
photon antibunching, photon emission, single-photon source, transdimensional plasmonic materials

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