1 Title

Energy transport and plasmon dispersion in metal-insulator-metal devices

2 Abstract

For increasing speed and bandwidth for on-chip communication it is very convenient to use light. However, present day photonic chips have a footprint of at least one square centimeter, whereas electronic components can already be build at the nanometer scale. This mismatch makes it critical for photonic component to be scaled down to the nanometer regime. One possiblity to do this, is by using plasmonics.

We propose a detailed study of a very promising plasmonic device: a Metal-Insulator-Metal (MIM) waveguide with an array of quantum dots embedded in the insulator. Using the Green's tensor formalism and the density-matrix approach, we will give an integrated description of exciting surface plasmons with quantum dots, transfering energy through an array of quantum dots and guiding plasmonic modes through a MIM waveguide. All this will be studied on the subwavelength scale.

Recently Walter et al [1] showed that it is possible to fabricate such a device that is compatible with current CMOS-technology. For further implementation and development of these devices it is of crucial importance to understand the important physical processes and the relevant parameters.

3 Applicants

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4 Institute

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5 Duration of the project

4 years, starting in september 2011

6 Personnel

6.1 Senior-scientists

J. Knoester, group leader and promotor, 10%
V. A. Malyshev, supervisor, 20%

6.2 Junior-scientists

P. J. Compaijen, OIO full time

7 Cost estimates

7.1 Personnel positions

1 OIO, four years, k€ 204.000

7.2 Running budget

4 × k€ 5.000 for standard expenses

7.3 Budget summary

	2011	2012	2013	2014	2015	Total
personnel (positions): PhD student	1/4	1	1	1	3/4	4
personnel (costs):	k€ 12.75	k€ 51	k€ 51	k€ 51	k€ 38.25	k€ 204
running budget	k€ 1.25	k€ 5	k€ 5	k€ 5	k€ 3.75	k€ 20
TOTAL (requested from FOM)	k€ 14	k€ 56	k€ 56	k€ 56	k€ 42	k€ 224

8 Research Programme

8.1 Introduction

8.1.1 Context: nanophotonic circuitry

Photonics describes generation, emission, transmission, processing and detection of light. It covers technical applications over the whole spectrum from ultraviolet to far-infrared. Over the past years photonics has played a major role in the development of high-speed communication. It is expected to be one of the key technologies and drivers for innovation in the 21st century [2]. Nowadays, the footprint of a photonic chip is at least one square centimeter. One of the greatest challenges for photonics will be the downscaling to nanometer sizes and the integration with nano-electronics. Using plasmons, which are collective excitations of free electrons in metals, it is possible to manipulate and guide light well below the diffraction limit [3]-[14]. Therefore, plasmonics is expected to

play a prominent role in this miniaturization. Plasmonic devices can be build with sub-wavelength dimensions and are therefore uniquely capable of bridging the size-mismatch between microphotonics and nanoelectronics and combine the best of both worlds [15].

8.1.2 The problem: guiding light at nanometer scale

Conventional integrated optical components use dielectric waveguides to manipulate light. However, these waveguides have a minimal bending radius of several millimeters, yielding chips with typical footprints of square centimeters (see [6] and references therein). Surface plasmon polaritons (SPPs), which are collective excitations of plasmons and photons, hold enormous potential for guiding light at the nanometer-scale. SPPs can be excited at the interface between a metal and a dielectric and can propagate along this interface. These SPPs are confined to the interface well below the excitation wavelength. However, since excitations of conduction band electrons in metal suffer from both interband relaxation and free-electron scattering, the propagation of SPPs is always damped. Typical values for the energy attenuation length are around 10 - 100 micron (in the visible region), depending on the metal-dielectric configuration. A typical trade-off for plasmonics is that the better the confinement to the interface is, the shorter the propagation length will be [3]. This implies that in order to be able to guide light efficiently on the nanometer-scale, one somehow needs to work around this trade-off. One of the possible solutions is guiding the plasmons along a chain of closely spaced metal nanoparticles ([12], [13] and [14]). In this way, each particle will excite new plasmons giving rise to SPP guiding over 120 micron [16]. However, in such systems the losses are still quite high. Another proposed method to increase the SPP propagation is by making use of different geometries. Especially the Metal-Insulator-Metal (MIM) waveguides are very promising configurations [17]. If the inter-spacing between the two metal plates is small enough, only plasmonic modes will be supported and therefore more energy will be coupled into these modes [18].

To integrate optics in nano-electronic chips, there is another complication: the local excitation of SPPs. Looking at the dispersion relation of SPPs (see Fig. 2), one can see that the SPP line lies to the right of the light line, which implies that this mode cannot be excited by a laser pulse. Rather, special phase-matching techniques are required (such as gratings or prisms [4]). Another possibility to excite SPPs is by making use of the near field of an oscillating dipole or excited quantum dot. The near field is the region closer to the dipole than the emission wavelength [19]. The modes present in this field are evanescent waves, and therefore have sufficient momenta to excite SPPs. In view of applications it is interesting to use quantum dots as emitters, since they can be excited electronically.

However, placing a quantum dot close to a metal will change the radiative and non-radiative decay properties of the dots. Also the coupling between individual quantum dots as well as their collective optical response in close proximity of a metal surface are still open questions. Furthermore, for both practical and fundamental purposes it is needed to study the dispersion relation of the plasmon modes excited by the quantum dots and quantify the propagation of these modes in the MIM waveguide.

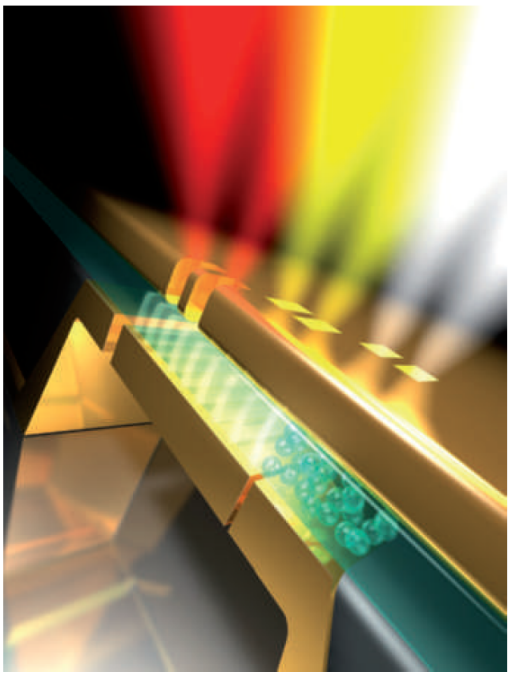

Figure 1: Artistic impression of a CMOS-compatible SPP source, fabricated by Walters et al [1]. Silicon nanocrystal (shown in green) emit SPPs into a MIM waveguide. The holes on the top metal contact can be used as output couplers. Reproduced from [15]

8.1.3 Aim: towards an integrated description of plasmons and quantum dots in MIM devices

The above reasonings of electronic SPP excitation using quantum dots, efficient energy coupling of quantum dots into plasmon modes for MIM devices and guiding SPPs by arrays of emitters, call for a detailed understanding of quantum dots in MIM devices, from both a functional and fundamental point of view. Specifically, we plan to study: (i) excitation of plasmons by quantum dots, (ii) energy transfer between quantum dots and (iii) propagation of plasmons in the presence of quantum dots, taking into account the mixing of plasmon-quantum dot eigenmodes.

8.2 Method

In this section, we present a method needed to obtain a full description of quantum dots embedded in Metal-Insulator-Metal waveguides. Since many different interactions play a role in the system under study, we will divide the system into several parts. It should be noticed that some of these parts have already been studied quite extensively. However, this project is unique in combining these different parts and arriving at a phenomenological description for understanding the excitation and propagation of plasmons in MIM devices.

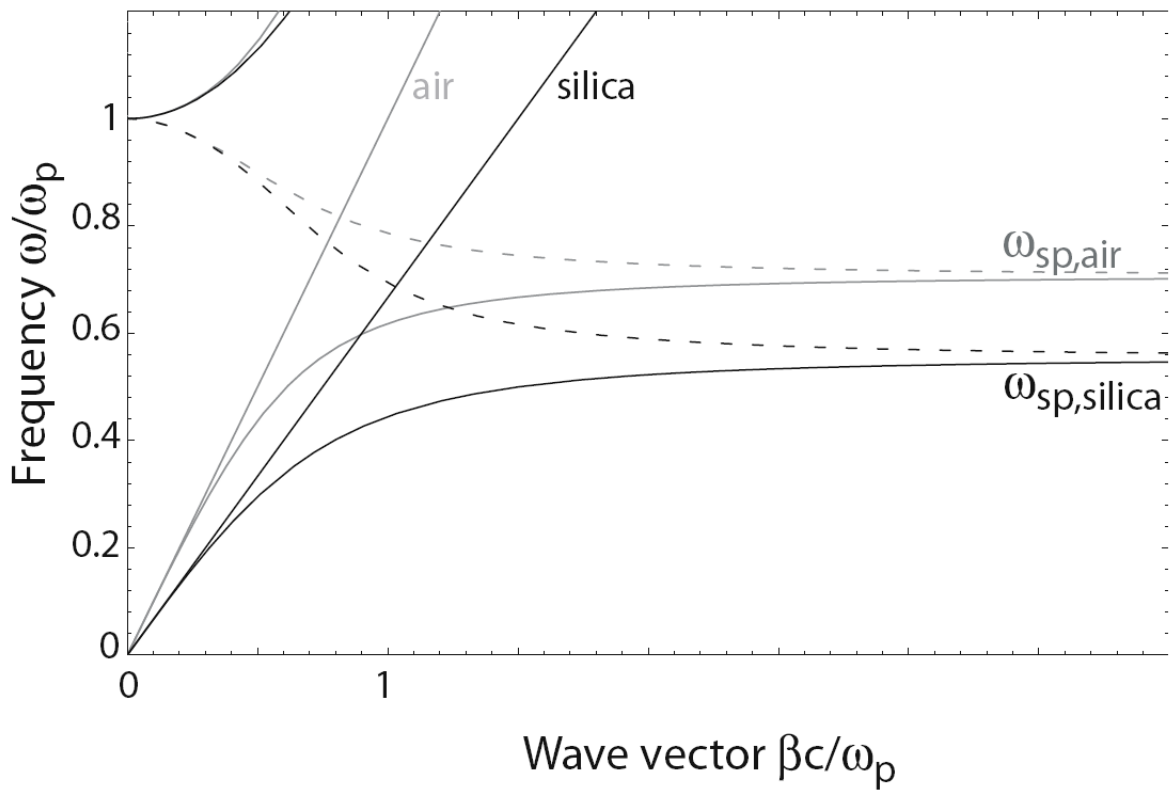

Figure 2: Dispersion relations for SPPs on the interface between a drude metal with negligible damping and air (gray) and silica (black). Fig. reproduced from [3]

8.2.1 A single quantum dot close to a metal surface

A quantum dot may be considered as a three-dimensional quantum well. The energy levels are quantized for all three directions of motion [20]. The confined excitons in such quantum dots have a high quantum efficiency for excitation and radiative decay. The efficiency and the band gap are determined by the semiconductor material used and the size of the dot. The band gap for a bulk semiconductor CdSe has a band gap in the infrared. However CdSe quantum dots with radii around 3 nm have their band gap in the visible [4].

If an oscillating species (molecule, metal nanoparticle, quantum dot) is much smaller than the excitation wavelength, the oscillator can be treated as a point dipole source. Under this assumption the problem of an emitter above a metal surface was first treated by Chance, Prock and Silbey (CPS), which they published in a series of papers, summarized in a classic review article [21].

The electric field at a certain position $\mathbf{r}$ radiated by three orthogonal dipoles at a source position $\mathbf{s}$, can be calculated using the Green's tensor formalism [26]. For a homogeneous medium, this tensor can be calculated analytically. However, when a surface is present, there will also be a scattered contribution to the Green's tensor:

$$\mathbf{G}(\mathbf{r},\mathbf{s},\omega) = \mathbf{G}^H(\mathbf{r},\mathbf{s},\omega) + \mathbf{G}^S(\mathbf{r},\mathbf{s},\omega) \tag{1}$$

where $\mathbf{G}$ is the full Green's tensor, $\mathbf{G}^H$ the homogeneous and $\mathbf{G}^S$ the scattered Green's tensor.

The Green's tensor for the scattered electric field can be determined using the method proposed by Sommerfeld in 1909 [25]. This method is based on expanding the spherical waves produced by an oscillating dipole into plane waves (the Sommerfeld identity). Using this, the Fresnel reflection on the interface can be calculated. The general result of this treatment is the Sommerfeld integral. Some care has to be taken with evaluating these integrals. Due to the mathematically awkward behavior these integrals need a special numerical treatment (more information about this can be found in [25] and [26]). One of the contributions in this integral is the singularity that is present for TM-polarized modes, which corresponds to the SPP contribution. Since the contribution of the singularity can be calculated analytically, this method allows for an analytical calculation of the electric field produced by the SPPs that are excited by an oscillating dipole.

Once $\mathbf{G}^s(\mathbf{r}, \mathbf{s}, \omega)$ is known, one can calculate the dipole self-interaction via the interface and how this alters the radiative and non-radiative decay rates. By taking the imaginary part of the product of the scattered electric field at the dipole position and the dipole moment, the power dissipation can be calculated for all in-plane wave vectors [21].

An extension to the CPS model was made by Ford and Weber [24]. They showed that for small separations between the dipole and the metal, the wavevector dependence of the dielectric functions needs to be taken into account, i.e. including electron-electron interactions.

Using this model, very nice results have been obtained by Kalkman et al [22]. In this paper, a theoretical and experimental study has been performed on the excitation by surface plasmons at a SiO_2/Ag interface by silicon quantum dots. The results are shown in Fig. 3. Spontaneous emission corresponds to the in-plane wave vectors smaller than k_d, which is the wave vector of the plane waves in the glass. Values larger than k_d correspond to the far-field plane waves and represent energy dissipation to the interface. A peak close to k_d corresponds to the energy dissipated by the surface plasmon modes. Note that the vertical axis has a logarithmic scale and a large portion of the total energy is put into the surface plasmon.

The model outlined above appears to be ideally suited for studying the emission properties of quantum dots close to a metal interface. However, some care has to be taken with modeling the quantum dot as a classical oscillator. To be precise, one should threat a quantum dot fully quantum mechanically, i.e. using the density matrix formulation. In principle, these equations form a nonlinear set, but in the weak interaction limit (very small population in the excited state), they can be linearized and then the equations for a classical dipole will be obtained exactly. However, under stronger excitation, the system becomes nonlinear and the classical treatment is no longer valid. Under these conditions, there is still very little theoretical and experimental knowledge about the treatment of a quantum dot close to a metallic interface.

8.2.2 A single quantum dot in a Metal-Insulator-Metal waveguide

The Green's tensor calculation outlined above can be easily extended to two interfaces. Instead of one, the oscillating dipole will now interact with two image dipoles (one in each metallic layer). Calculations of the electric fields of an oscillating dipole in MIM

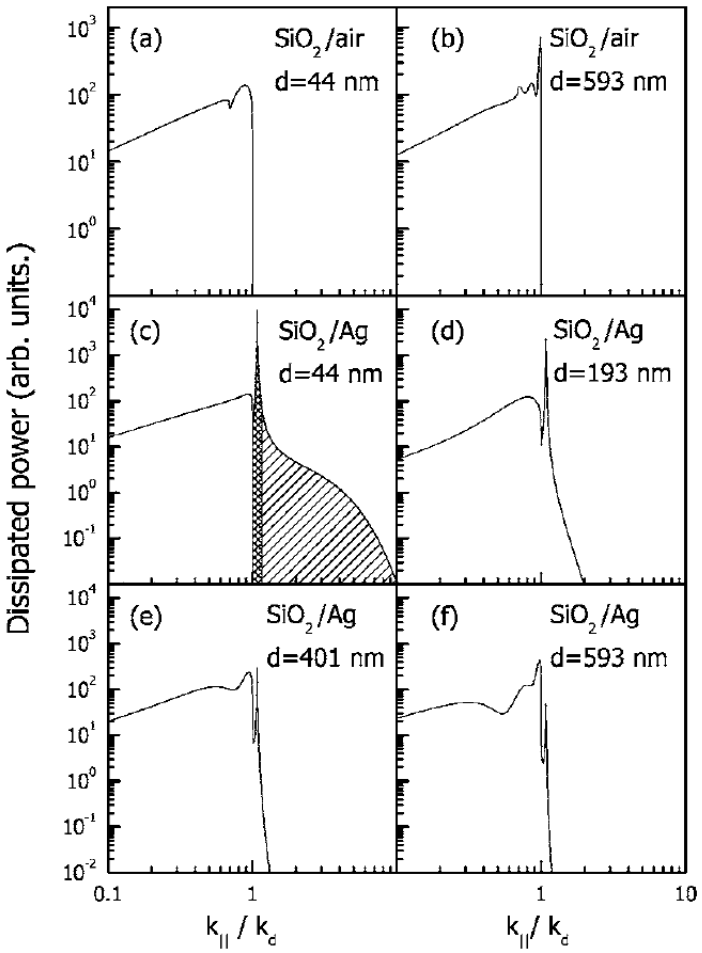

Figure 3: Power dissipation as a function of the in-plane wave vector component for a randomly oriented dipole located near a SiO_2/air and SiO_2/Ag interface at several heights. The quantity on the horizontal axis is the in-plane-wavevector normalized to the wavevector in glass. Fig. reproduced from [22]

waveguides ([18] and [21]) and general stratified media ([26]) have already been performed. Also, according to [27], the Sommerfeld integral loses a highly oscillatory part, which makes the convergence faster than for a single interface.

A very important feature of MIM waveguides is that the mode structure depends on the distance between the metallic plates. This means that if the spacing is small enough (less than 150 nm [1]) only bound surface modes can be present in the waveguide and therefore this is the only channel through which the quantum dot can lose its energy. Jun et al [18] reported a strong emission enhancement of the quantum dot, depending on the emission wavelength and the separation of the plates. These results are shown in Fig. 4.

8.2.3 Interaction between two quantum dots close to a metal surface

As was discussed in the previous sections, the spontaneous emission properties of a quantum dot can be altered by the presence of a surface, because the dot will interact with itself via the surface. A similar phenomenon will occur for the energy transfer between

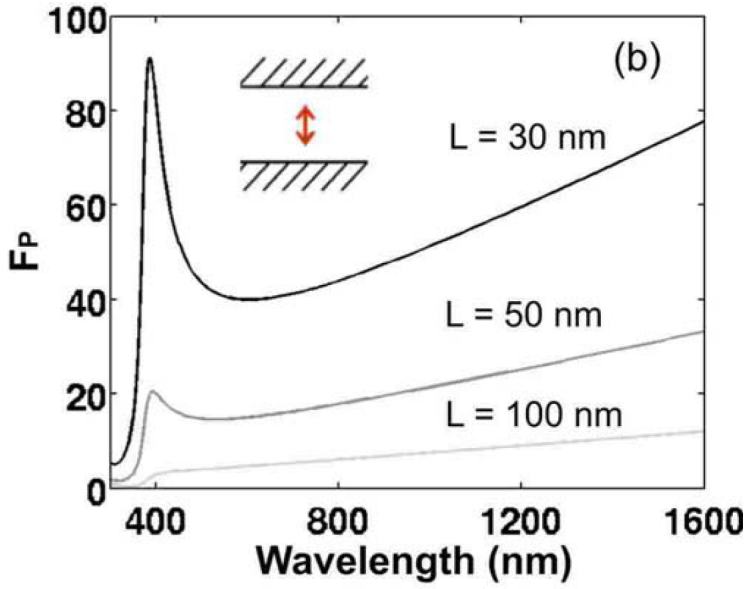

Figure 4: Emission enhancement factor of a MIM waveguide as a function of the emission wavelength and for different metallic plate separations. Fig. reproduced from [18]

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to quantum dots, because the electromagnetic field felt by the dots will be different from that in free space.

The principal mechanism for energy transfer between neighboring quantum dots is resonance energy transfer (RET). In the systems we plan to study, the separation R between the quantum dots will be much smaller than the wavelength of the emitted light and therefore we will use Försters theory for radiationless short-range energy transfer [28]. This type of transfer exhibits the R^{-6} dependence typical for Coulomb coupling between two dipoles, whereas the radiative energy transfer (dominating for separations much larger than the wavelength) behaves as R^{-2}. The full energy transfer theory can be described using quantum electrodynamics (see [29] and references therein).

A clear recipe for calculating this effect is given by Dung et al in ref. [30]. In their paper a rigorous derivation of the rate of intermolecular energy transfer in the presence of an arbitrarily shaped, dispersing and absorbing material is given. The starting point is a quantized version of the macroscopic electromagnetic field. The equation that they obtained for the energy transfer rate between two molecules situated at positions $\mathbf{r}_A$ and $\mathbf{r}_B$ is

$$\tilde{w}(\omega) = \frac{2\pi}{\hbar^2}(\frac{\omega^2}{\epsilon_0 c^2})^2 |\mathbf{d}_B^* \mathbf{G}(\mathbf{r}_B, \mathbf{r}_A, \omega)\mathbf{d}_A|^2 \tag{2}$$

where ω is the frequency corresponding to the emitted radiation, $\mathbf{d}_{A,B}$ are the dipole operators of the molecules at positions A and B respectively and $\mathbf{G}$ is the full Green's tensor taken at the positions of the two molecules. Close to the interface the contribution of the metal will be large and therefore the full Green's will be very different from the one in the homogeneous case. Hence, the influence of the surface on the energy transfer properties will be substantial. Although the derivation was performed for molecules, the exact same treatment can be used for quantum dots.

At this point it is interesting to make a comparison with the spontaneous decay rate of a single quantum dot. That property depends on the imaginary part of the Green's tensor calculated at the point of the dipole, whereas the energy transfer rate depends on the full two-point Green's tensor. This implies that the presence of the surface can affect both properties quite differently. Dung et al. even have observed situations in with the spontaneous decay was enhanced and the energy transfer was inhibited. Furthermore, it was shown that surface-guided modes have a large influence on the energy transfer.

8.2.4 An array of quantum dots close to a metal surface

In 2007, Gartstein and Agranovich published their research on the decay of excitons in long molecular chains near a reflecting interface [31]. The interaction of an excitation in a chain of identical molecules with a metal surface will lead to delocalization of the excitation and modification of the radiative decay. They observed that surface plasmons are an efficient decay channel for one-dimensional excitons. The method they used for solving this problem is by assuming a very long chain of molecules and excitons that are completely delocalized over this chain. The interaction of the exciton with itself via the interface is again calculated using Sommerfelds treatment. This method allows for a quite straightforward calculation of the decay properties of a delocalized exciton in a molecular chain. However, since a delocalized exciton is considered, only the collective response of the molecules can be calculated.

The same treatment can be used for describing the collective response of an array of quantum dots close to the metal. However, to understand the local excitation and guiding of the surface plasmons, we need the local response of each emitter to the excitation. This problem can be studied in the dipole approximation using the methods described in the previous sections. For continuous wave excitation of the first dot we can calculate how the excitation decays over the chain and over the surface. When the full array is excited, one has to study how the exciton is delocalized over the chain and assign a dipole moment to each emitter relative to the probability of finding the exciton at that position. This way an approximation of the excitation of SPP modes by each particle can be given. How good this approximation will be and how a more extensive treatment can be performed has to be investigated in more detail.

8.3 Applications: systems of special interest

The methods described above allow for studying a wide variety of systems. For this project the focus is on local excitation of surface plasmons and the guiding surface plasmon efficiently at a subwavelength-scale. The final goal is to give an integrated description of SSP creation and guiding by an array of quantum dots in a metal-insulator-metal device as was recently fabricated by Walters et al [1]. Although the systems described below are in itself interesting and important, they will also be considered as steps towards the final goal.

8.3.1 Decay properties of a single quantum dot close to a metal surface under strong optical and electronic excitation

As was discussed in section 8.2.1, the decay properties of emitters in the vicinity of a reflecting interface have already been studied for over thirty years. One thing in common in all these studies is that the emitting species is modeled as a classically oscillating dipole. Although this is a good approximation under the assumption of weak excitation, it fails to describe a quantum dot when the excitation becomes stronger. Under these conditions the density matrix approach has to be used. Recently, interesting behavior has been observed by Artuso et al ([32],[33]) for quantum dots-metal nanoparticle dimers under strong excitation. The interaction of the quantum dot with its image (induced into the metal nanoparticle) gives rise to a bistable optical response of the dimer. This bistable response allows for optical switching of the system. A physical property that is very important for the applicability of this system as an optical switch is the decay time of the quantum dot. This parameter will determine how fast the system can be switched. In this subtopic we will investigate if the same behavior can be observed for a quantum dot close to a metal surface or embedded in a MIM geometry and if so, what the corresponding switching time will be. Also, we will study if the interaction of the quantum dot and the metal changes when using electronic excitation and how this should be treated mathematically.

8.3.2 Energy transfer and propagation in an isolated array of quantum dots

Although the isolated array is not related to excitation of SPPs nor to the guiding of these modes, understanding the fundamental interactions present in an array of quantum dots is of critical importance for studying the array embedded in MIM waveguide. When considering weak optical excitation, the solution to this problem is comparable with calculating the spectrum of a linear aggregate of identical molecules (e.g. [31]). However, under stronger excitation, multiple excitons will be created in the array and the interactions between these excitons need to be taken into account. The importance of understanding these interactions is that when the array is embedded in a MIM device, the dots will be excited by applying a voltage over both metal cladding layers. Because of this voltage, a current will run through the isolating layer and excite the quantum dots. In general this means that more quantum dots will be excited and the created excitations will interact.

Furthermore, it will be interesting to study this system in the nonlinear regime. As becomes clear from [34] and [35], there is some discussion about how to calculate the optical response of linear aggregates in under nonlinear conditions. The usual treatment is to split the global density matrix into a product of one-molecule density matrices. For long aggregates this leads to a bistable response. However, for a dimer is was shown that this non linear response is merely an artifact of the calculation. For molecular aggregates it is difficult to verify this result experimentally, since the experiment has to be performed at the single aggregate scale and the disorder has to be very low. However, for quantum dots it is nowadays easily possible to create a single array with small disorder. This allows for a verification of this commonly used theoretical model.

8.3.3 Propagation and dispersion of SPPs excited by an array of quantum dots in a MIM waveguide

The final system under study is the Metal-Insulator-Metal waveguide with quantum dots embedded in the insulator. This system is interesting to study, because of the high potential it has for facilitating on-chip creation and guiding of SPPs. In order to be able to calculate the SPP propagation and dispersion, we need to combine the methods and subprojects described above. This treatment allow for a detailed study of the origin and nature of the SPP modes that were observed by Walters et al. Also we can investigate how the SPP modes couple to the eigenmodes of the quantum dot array.

An integrated description of this system is of critical importance for understanding and developing optical on-chip communication. The influence of the dielectric properties of the used materials, the separation between the two metal plates, the excitation strength and the emission wavelength of the quantum dots will be studied.

8.4 Workplan

Months 1-6: Studying a single quantum dot close to a metal surface.

Months 7-12: Calculating the interaction between two quantum dots close to a metal surface.

Months 13-24: Treating an isolated array of quantum dots

Months 25-45: Investigating the propagation and the dispersion of SPPs excited by arrays of quantum dots close to a metal surface and embedded in a metal-insulator-metal waveguide.

Months 46-48: Finishing thesis based on publications.

8.5 Embedding of the proposed research

The research will be carried out in the group Theory of Condensed Matter, headed by prof. dr. Jasper Knoester. The daily supervision will be done by dr. Victor. A. Malyshev. In this research group several projects on plasmonics have been carried out already, e.g. energy transport in graded arrays of metal nanoparticles, guiding of SSPs by arrays of metal nanoparticles and the coupling between metal nanoparticles and semiconductor quantum dots. Outside the institute we will collaborate strongly with the Polman group, where recently MIM devices with quantum dots have been fabricated [1].

9 Application perspective in industry, other disciplines or society

As was pointed out in the introduction, plasmonics is expected to be the key technology for driving innovation in the 21st century. Exciting and guiding plasmons at the nanometer scale will lead to miniaturization of photonics and therefore will yield a huge

cost reduction per bit. Also, it will be possible to add new functionality to integrated circuitry. For developing these technologies, not only the scientific component of this research is important, but a crucial role will be played by small and medium enterprises [2]. This implies a great benefit for the Netherlands when taking part in this research. However, the proposed research is not only interesting from a functional point of view. The ability to describe the fundamental excitations that can be present in the devices under study and the possibility to find new mixed plasmon quantum dot eigenmodes will contribute greatly to the understanding of electrodynamics close to reflecting interfaces.

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