# The Simulation Scenario of Quantum Tunneling in Gold Nanodimers within a Classical Framework

**Mandana Jalali[a], Jan Taro Svejda[a], Jesil Jose[b], Sebastian Schlücker[b], Daniel Erni[a]**

[a]*General and Theoretical Electrical Engineering (ATE), Faculty of Engineering, University of Duisburg-Essen, and CENIDE – Center for Nanointegration Duisburg-Essen, 47048 Duisburg, Germany*
[b]*Department of Chemistry, University of Duisburg-Essen, and CENIDE – Center for Nanointegration Duisburg-Essen, Universitätsstr. 5, 45141 Essen, Germany*

The quantum tunneling within the gap between two gold nanowires is studied in order to apprehend the effect of wall's curvature on the effective quantum tunneling volume. To accurately model the gold dimer with sub-nanometer gap sizes, the hydrodynamic model together with the quantum corrected model are implemented within a fully classical finite element method (FEM) solver, which has facilitated the simulation of both large nanodimers as well as large gaps up to 2nm. Results show that as the wall's curvature determines the effective tunneling volume, the gap sizes in which the quantum tunneling starts is dependent on the nanodimer's sizes.

## INTRODUCTION

Concentrating light in sub-diffraction volume has many applications, and hence has been extensively studied based on the Maxwell equations [1]. However, it is shown that when the size of the nanoparticle becomes in the range or smaller than the finite Fermi wavelength of electrons, this classical picture can no longer fully describe the optical response of the nanoparticle as the permittivity of the nanoparticle as well as being frequency dispersive is non-local and thus depend on the wave vector [2, 3]. Such nonlocality has been described in various models such as the hydrodynamic approach or the generalized nonlocal optical response (GNOR) model [4], which has been implemented to realize the optical response of small plasmonic nanoparticles [5, 6]. Nevertheless, when the gap sizes in metallic dimers becomes in subnanometer range where the quantum tunneling starts to occur, these models fail to provide a full description of the dimer's optical response and hence a full quantum mechanical description of the structure is required which can only be done for small dimers [7, 8]. An alternative approach which facilitates studying large dimers as well as large gap sizes up to 2nm, is to translate the quantum tunneling probability into a fictitious DC conductivity of the gap in the framework of the quantum corrected model (QCM) in order to bridge quantum and classical plasmonics [9, 10]. In this work, the hydrodynamic model as well as the quantum corrected model are implemented within a classical finite element solver, in order to investigate the effect of the wall's curvature on the effective quantum tunneling volume in gold nanowire dimers with various radii and gap sizes.

## FICTITIOUS MATERIAL

In the quantum corrected model, the electron transfer probability within the gap $T(l)$ is converted into a static conductivity and a fictitious material is defined for the gap with the mentioned DC conductivity [11]. This DC conductivity is dependent on the nanowires separation distances and consequently on the nanowires curvature. The Drude permittivity

of such gaps between two gold walls is shown in the equation (1).

$$\varepsilon_g(\omega,l) = \varepsilon_0(\omega) + (\varepsilon_m^d(\omega) - \varepsilon_0(\omega))e^{-l/l_d} - \frac{\omega_P^2}{\omega(\omega + i\gamma_g(l))} \tag{1}$$

In which the $\gamma_g(l) = \gamma_P e^{-l/l_c}$ is the separation-dependent loss parameter with the phenomenological decay length of $l_c = 0.4$Å. The second term in the equation (1) accounts for the contribution of the d-electrons to the optical response of the dimer, in which the dispersive $\varepsilon_m^d$ associates the interband transitions involving d-electrons with the decay length of $l_d = 0.8$Å to the consequent conductivity. The real part of the fictitious permittivity together with the corresponding conductivity are illustrated in the Figure 1 b), c) for a 15nm with a 0.2nm gap size for various energies along the gap width (the red dashed line within the Figure 1 a). As depicted, both the conductivity and the real part of the permittivity are dispersive and decay rapidly as the separation distance increases. The conductivity is presented for various gap sizes in both 15nm and 40nm dimers at the $E = 3.1$eV (cf. Figure. 1 d). The interesting feature of this graph is that although the magnitude of conductivity is the same for both dimers the effective conductivity volume is larger in the larger dimer and hence the resulted quantum tunneling current should be dependent on the dimer's size. This feature is better presented via the conductivity profile for both dimers in the Figure 1 e), f).

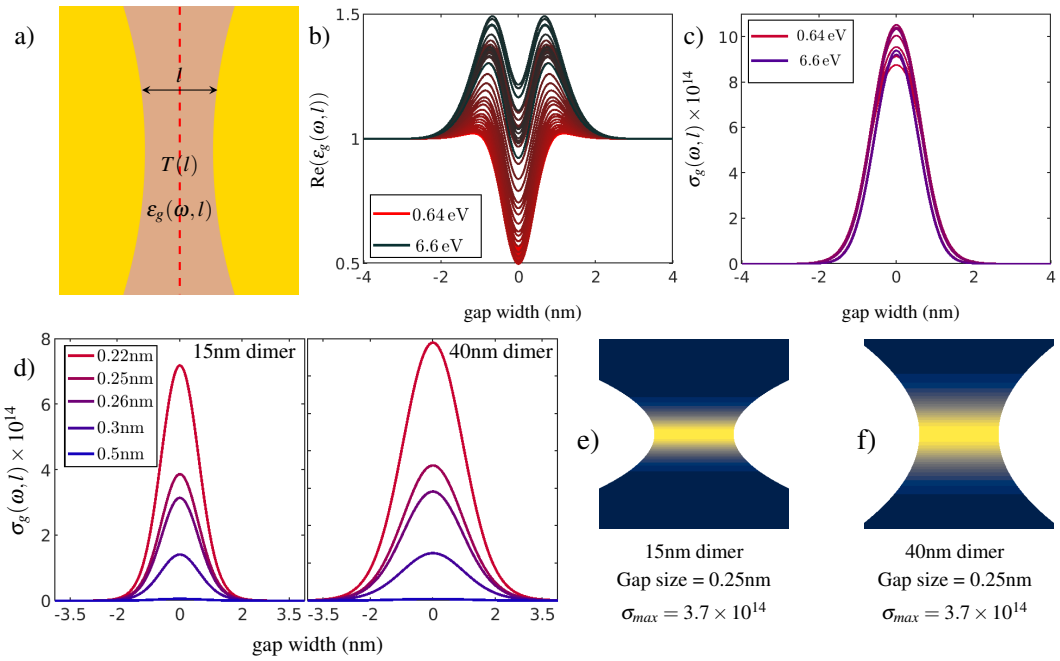


Figure 1: a) The schematic of the nanodimer geometry where the red dashed line depicts the gap width. b) The real part of the fictitious material permittivity for various energies along the gap width. c) The gap conductivity for various energies along the gap width. d) The gap conductivity for different gap sizes in both 15nm and 40nm dimers along the gap width together with the gap's conductivity profiles for e) 15nm and f) 40nm dimers.

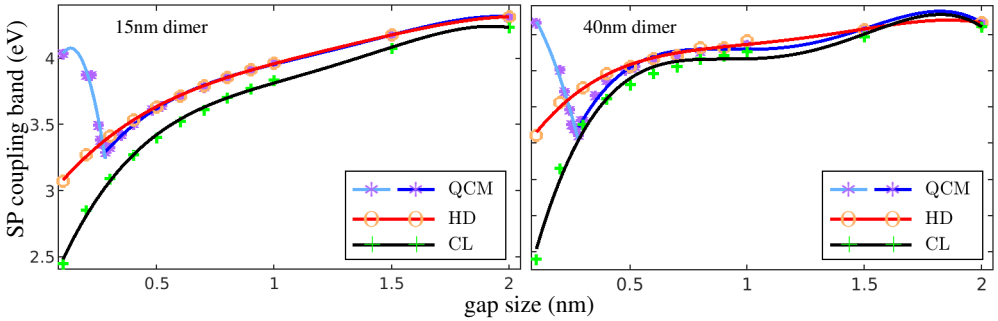


Figure 2: The SP coupling band in both 15nm and 40nm dimers versus the gap sizes based on the quantum corrected model (QCM), the hydrodynamic model (HD) as well as the classical model (Cl), where the points are the simulation results which are then fitted to a polynomial (solid lines).

## MODELLING AND SIMULATION SCENARIO

The modelling is carried out in a classical FEM solver namely Comsol Multiphysics in which the hydrodynamic system of equations is defined as partial differential equations and added to the Maxwell equations [12]. The hydrodynamic current's normal component is set to zero on the nanowires and the gap boundaries, which ensures that the hydrodynamic current only exists within the mentioned regions. The QCM model is added to the gap via the represented DC conductivity where separation distance dependency is taken into account. To reduce the required computational resources as well as the simulation time, the geometry is cut vertically into half (along the red dashed line in the Figure 1 a)), while adding the appropriate boundary conditions. A fine meshing is required to accurately resolve the subnanometer gap which results in 453091 degrees of freedom even for the smallest dimer. The surface plasmon (SP) coupling band is determined as the main figure of merit for 15nm and 40nm gold dimers with gap sizes ranging from 0.1nm up to 2nm based on the fully classical model, the hydrodynamic model as well as the QCM model.

Table 1: The average hydrodynamic conductivity at corresponding SP coupling bands in 15nm and 40nm dimers for various gap sizes.

| | | 0.2nm | 0.3nm | 0.4nm | 0.5nm |
|---|---|---|---|---|---|
| SP coupling band eV | 15nm | 3.79 | 3.32 | 3.51 | 3.64 |
| | 40nm | 2.40 | 2.26 | 2.37 | 2.41 |
| $\sigma_{hd}\ \frac{\text{kS}}{\text{m}}$ | 15nm | 28.9 | 3.2 | 0.28 | 0.02 |
| | 40nm | 84.8 | 6.9 | 0.57 | 0.05 |

## THE QUANTUM TUNNELING CURRENT

The SP coupling band for all the gaps in each model is fitted to a $4^{\text{th}}$ order polynomial to study the spectral position of the SP coupling bands (cf. Figure 2). As expected the deviation of the hydrodynamic model from the classical model starts in larger gap sizes for the 15nm dimer, as the hydrodynamic effect is stronger in smaller nanoparticles. However,

the deviation of the QCM model from the hydrodynamic model starts earlier in the larger dimer. This deviation is the result of the quantum tunneling within the gap and shows that quantum tunneling starts at larger gap sizes for larger dimers compared to the smaller ones. This effect is the consequence of the larger effective conductivity volume in larger dimers due to a more gradual curvature. In order to validate this claim, the average hydrodynamic conductivity (AHDC) of the gap $\sigma_{hd} = \int_{gap} \frac{J_{hd}}{|E|}$ at the SP coupling band is calculated for both dimers in gap sizes in which the quantum tunneling occurs. The results are presented in table 1, which shows the larger AHDC in larger dimers and consequently a size-dependent red shift in the SP coupling band.

# References


[1] S. A. Maier, *Plasmonics: fundamentals and applications* (Springer Science & Business Media, 2007).

[2] M. S. Tame, K. R. McEnery, Ş. K. Özdemir, J. Lee, S. A. Maier, and M. S. Kim, "Quantum plasmonics," Nat. Phys. **9**, 329-340 (2013).

[3] Y. Luo, A. I. Fernandez-Dominguez, A. Wiener, S. A. and Maier, and J. B. Pendry, "Surface plasmons and nonlocality: a simple model," Phys. Rev. Lett. **111**, 093901 (2013).

[4] S. Raza, S. I. Bozhevolnyi, M. Wubs, and N. A. Mortensen, "Nonlocal optical response in metallic nanostructures," J. Phys. Condens. Matter **27**, 183204 (2015).

[5] J. M. McMahon, S. K. Gray, and G. C. Schatz, "Calculating nonlocal optical properties of structures with arbitrary shape," Phys. Rev. B **82**, 035423 (2010).

[6] F. J. Garcia de Abajo, "Nonlocal effects in the plasmons of strongly interacting nanoparticles, dimers, and waveguides," J. Phys. Chem. C **112**, 17983–17987 (2008).

[7] J. Zuloaga, E. Prodan, and P. Nordlander, "Quantum description of the plasmon resonances of a nanoparticle dimer," Nano Lett. **9**, 887–891 (2009).

[8] T. V. Teperik, P. Nordlander, J. Aizpurua, and A. G. Borisov, "Quantum effects and nonlocality in strongly coupled plasmonic nanowire dimers," Opt. Express **21**, 27306–27325 (2013).

[9] R. Esteban, A. G. Borisov, P. Nordlander, and J. Aizpurua, "Bridging quantum and classical plasmonics with a quantum-corrected model," Nat. Commun. **3**, 1–9 (2012).

[10] W. Zhu, R. Esteban, A. G. Borisov, J. J. Baumberg, P. Nordlander, H. J. Lezec, J. Aizpurua, and K. B. Crozier, "Quantum mechanical effects in plasmonic structures with subnanometre gaps," Nat. Commun. **7**, 1–14 (2016).

[11] R. Esteban, A. Zugarramurdi, P. Zhang, P. Nordlander, F. J. García-Vidal, A. G. Borisov, and J. Aizpurua, "A classical treatment of optical tunneling in plasmonic gaps: extending the quantum corrected model to practical situations," Faraday Discuss. **178**, 151–183 (2015).

[12] G. Toscano, S. Raza, A. Jauho, N. A. Mortensen, M. Wubs, "Modified field enhancement and extinction by plasmonic nanowire dimers due to nonlocal response," Opt. Express **20**, 4176–4188 (2012).