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Classical and Quantum Theory of Nanoparticles

At the nanoscale, materials behave differently from how they generally do. The surface effects are not negligible anymore and it is not possible to completely rely on the classical laws of Mechanics and Electromagnetics. Nanotechnologies represent a bridge between the macroscopic daily life world and the microscopic reality governed by the strange quantum laws. These structures are so small that cannot be *touched* so easily and often the only way to control and interact with them is by light. That's why the Scattering Electromagnetic Theory plays a key role in the multicolored and astonishing landscape of Nanotechnologies. In particular, Plasmonics is one of the main pillars of the building, and the collective electron motion in noble metals and in graphene is one of the most promising platforms to enhance the light-matter interaction effects. Our research group studies the electromagnetic properties of dielectrics, metals and semimetals at the nanoscale. We deal with arbitrary shape and size nanostructures in the full wave regime, providing both classical and quantum results. We perform theoretical and numerical analysis of the systems under investigation, trying to understand what the best ways to tackle the scattering problem are, in order to optimize the analysis and the design of custom nanostructures for photonic applications.

In these past two years of my PhD, I was involved in all the activities of our research group. My personal main interest mostly lied in the quantum analysis of localized plasmon nanoparticles, in the understanding of their damping mechanisms and in the calculation of the radiative lifetimes and of the frequency shifts of the plasmon modes, as the dimension of the nanoparticle varies. A brief sketch of the basic electro-fluid-dynamic formulation of the scattering problem by plasmon nanoparticles and of its quantum counterpart is given below.

Quantum Electrodynamical formulation of the scattering problem by metal nanoparticles

We consider a jellium model for the electron gas inside a metal nanoparticle. In the hypothesis of incompressible motion, the induced charges are confined on the surface of the particle. By fixing the Coulomb gauge for the vector potential $\nabla \cdot \mathbf{A} = 0$, saying $\xi(\mathbf{r}, t)$ the displacement electron field, which is null outside the particle domain Ω , and $\xi_n(\mathbf{r}, t)$ its normal component on the boundary $\partial\Omega$, the Lagrangian of the system can be written as follow:

$$L = \int_{\Omega} \frac{1}{2} mn_0 |\dot{\xi}|^2 - \frac{e^2 n_0^2}{8\pi\epsilon_0} \oint_{\partial\Omega} \frac{\xi_n(\mathbf{r})\xi_n(\mathbf{r}')}{|\mathbf{r} - \mathbf{r}'|} + \int_{R^3} \left[\epsilon_0 (\partial_t \mathbf{A})^2 - \frac{1}{2\mu_0} (\nabla \times \mathbf{A})^2 \right] - \frac{en_0}{c} \int_{\Omega} \dot{\xi} \cdot \mathbf{A}$$

n_0 = Electron density at rest
 c = Light velocity in vacuum
 e = Electron charge
 m = Electron mass

By expanding the displacement field into a set of curl and divergence free elements $\xi(\mathbf{r}, t) = \sum_m q_m(t) \mathbf{U}_m(\mathbf{r})$ and by performing the Legendre transformation with respect to the conjugate momenta $p_m = \partial L / \partial \dot{q}_m$, $\Pi = \delta L / \delta \dot{\mathbf{A}}$, we derive the following Hamiltonian:

$$H = \sum_m \frac{1}{2M_m} [(p_m + en_0 \langle \mathbf{U}_m, \mathbf{A} \rangle_{\Omega})^2 + M_m^2 \Omega_m^2 q_m^2] + \int_{R^3} \left[\frac{\Pi^2}{2\epsilon_0} + \frac{(\nabla \times \mathbf{A})^2}{2\epsilon_0} \right]$$

$M_m = n_0 m V_m$ Plasmon mass
 $V_m = \int_{\Omega} |\mathbf{U}_m(\mathbf{r})|^2$ Plasmon volume
 Ω_m = Plasmon EQS Resonance

This classical reformulation of the problem enables its canonical quantization:

$$H = \sum_m \hbar \Omega_m a_m^{\dagger} a_m + \int_{R^3} \frac{d\mathbf{q}}{(2\pi)^3} \sum_{s=1,2} \hbar \omega_q a_{q,s}^{\dagger} a_{q,s} + \sum_m \int_{R^3} \frac{d\mathbf{q}}{(2\pi)^3} \sum_{s=1,2} [V_{q,s}^m (a_m^{\dagger} - a_m) a_{q,s} + h.c.] + \sum_m \int_{R^3} \frac{d\mathbf{q}}{(2\pi)^3} \int_{R^3} \frac{d\mathbf{q}'}{(2\pi)^3} \sum_{s,s'=1,2} [W_{q,q'}^{s,s'} a_{q,s} a_{q',s'} + G_{q,q'}^{s,s'} a_{q,s} a_{q',s'}^{\dagger} + h.c.]$$

$$V_{q,s}^m = \frac{i\hbar\omega_p}{2} \sqrt{\frac{\Omega_m}{V_m\omega_q}} U_{q,s}^*(\mathbf{q}) \quad W_{q,q'}^{s,s'} = \frac{\hbar e^2 n_0}{4\epsilon_0 m} \frac{1}{\sqrt{\omega_q \omega_{q'}}} \eta(\mathbf{q} + \mathbf{q}') \mathbf{e}_{q,s} \cdot \mathbf{e}_{q',s'} \quad G_{q,q'}^{s,s'} = \frac{\hbar e^2 n_0}{4\epsilon_0 m} \frac{1}{\sqrt{\omega_q \omega_{q'}}} \eta(\mathbf{q} - \mathbf{q}') \mathbf{e}_{q,s} \cdot \mathbf{e}_{q',s'}$$

η = Characteristic function of the set Ω
 $f(\mathbf{q}) = \int_{R^3} f(\mathbf{r}) e^{-i\mathbf{r} \cdot \mathbf{q}}$

Currently, I'm spending a research period at The Institute of Photonic Sciences (ICFO) in Castelldefels (Barcelona, Spain) under the supervision of Prof. Darrick Chang, one of the most expert scientists of the world in the field of Atomic Physics. I hope to learn how to study atom arrays from his huge experience and to start a fruitful collaboration with him on the study of dipole-dipole quantum interaction in exotic environments.

One of the main issues of storing information in atoms and molecules is the decoherence. Even if we consider a quantum emitter in the free space, it will never constitute an isolated system, because of the always present vacuum oscillations. Quantum emitters interact with the continuous of radiative electromagnetic modes, losing coherence and energy in time. By the way, two great phenomena deserve attention. The first one was pointed out by Edward Mills Purcell in 1946, when he noticed that the spontaneous emission rate of an atom is not one of its intrinsic properties but it does depend on the surrounding environment. The second one has been highlighted very recently by the Quantum Optics community: the dipole-dipole interaction between quantum emitters is not negligible and it can dramatically change the spontaneous emission rate and the resonance frequency of the single atom.

In my last PhD year, I'd like to get into this problem of quantum memories, studying how we can use plasmon and dielectric nanostructures to control the behaviour of isolated quantum emitters and of quantum emitters arrays, in order to reach the best coupling coefficients to write and read information, perfectly subradiance state, and perfect reflection. From a theoretical point of view, this coupling problem is really interesting because the interaction between two open systems has to be modelled. The Hamiltonian is not hermitian and thus the time evolution is not a unitary group anymore.