Nonlocal Hydrodynamic Models for the Optical Response of Plasmonic Nanostructures

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***Abstract*—In order to model the interaction between light and plasmonic structures at deep-nanometer scale, which is governed by non-classical effects, a nonlocal hydrodynamic approach has been extensively studied. Several hydrodynamic models have been proposed, solving the coupled equations: the linearized hydrodynamic equation of motion and the electrodynamic Maxwell's equations, by employing additional boundary conditions. This work compares four hydrodynamic models: the hard wall hydrodynamic model (HW-HDM), the curl-free hydrodynamic model (CF-HDM), the shear forces hydrodynamic model (SF-HDM), and the quantum hydrodynamic model (Q-HDM). The analysis is conducted for a metallic spherical nanoparticle, as an example. The above hydrodynamic models are also compared with experiments available in literature. It is demonstrated that HW-HDM and Q-HDM outperform the other two hydrodynamic models.**

Keywords—additional boundary condition, deep-nanometer scale, nonlocal hydrodynamic model, plasmonics.

I. Introduction

Many conventional microwave topologies, such as a sphere, dipole, microstrip patch antenna, etc., have been successfully treated by using Maxwell's equations. However, the optical counterparts of these topologies, particularly the ones with deep-nanometric features, e.g., a metallic particle with a size of only a few nanometers, nanoparticle-on-mirror structure with a sub-nanometer gap, cannot be fully characterized by the abovementioned approach, due to the non-classical effects [1]. Notably, these effects may greatly affect the electromagnetic (EM) properties of the structure-under-study, which has been demonstrated in several plasmonic applications [2].

In order to study the non-classical effects, several semiclassical models, such as the hydrodynamic model (HDM) [3], have been introduced. HDM essentially investigates a multiphysics problem, namely the interaction between the mechanical motion of the free electron gas in metals and external EM fields. Metals are described by spatially dispersive (nonlocal) material parameters, meaning that the material response at a specific spatial point depends on the field in close vicinity of that point. On top, the material model introduces an additional wave, namely the longitudinal wave. In order to account for the longitudinal wave, HDM requires an additional boundary condition (ABC), in this way bridging the computational gap between the classical and *ab initio* quantum mechanical approaches.

However, the choice of HDM and ABC is not universal. Based on different physical approximations, several hydrodynamic models and ABCs have been proposed. Additionally, it has been demonstrated that different hydrodynamic models and ABCs may drastically modify the response of metallic nanotopologies [4,5].

This work reports on a comparison of the following hydrodynamic models: the hard wall hydrodynamic model (HW-HDM) with the Sauter ABC, the curl-free hydrodynamic model (CF-HDM) with the Pekar ABC, the shear forces hydrodynamic model (SF-HDM) with the specular reflection ABC, and the quantum hydrodynamic model (Q-HDM) with the corresponding ABC. The study is performed for a metallic nanosphere. For detailed discussion of the hydrodynamic models and ABCs, we refer the readers to our previous work [4].

II. Theoretical Framework

The studied structure and excitation are depicted in Fig. 1 (a). Gold is used as a constitutive metal, and glass as a surrounding medium. First, a hydrodynamic Drude model is employed, considering the contribution of free electrons. Then, the Drude model is extended by including the contribution of bound electrons. This is particularly important at optical frequencies, where bound electrons may considerably affect the response of noble metals [6]. The nanosphere is illuminated by an x-polarized plane wave, propagating along the z-axis.

In order to tackle the associated scattering problem, we implemented Mie's theory with spatial dispersion [7,8] for all hydrodynamic models. This approach expands the fields outside and inside the sphere into vector spherical harmonics. By applying the classical boundary conditions, which are augmented by ABCs, one may calculate the scattering coefficients [4].

III. Numerical Results

The following parameters for the permittivity of gold are used in the simulations: the plasma frequency $\hbar\omega_P$=9 eV, the damping frequency $\hbar\gamma$=0.05 eV, and the Fermi velocity v_F=1.4·10^6 m/s. The permittivity of the surrounding medium is 2.3.

The response of the nanoparticle is studied for both Drude and experimentally measured material models. The results for the extinction cross section are depicted in Figs. 1 (b) and (c).

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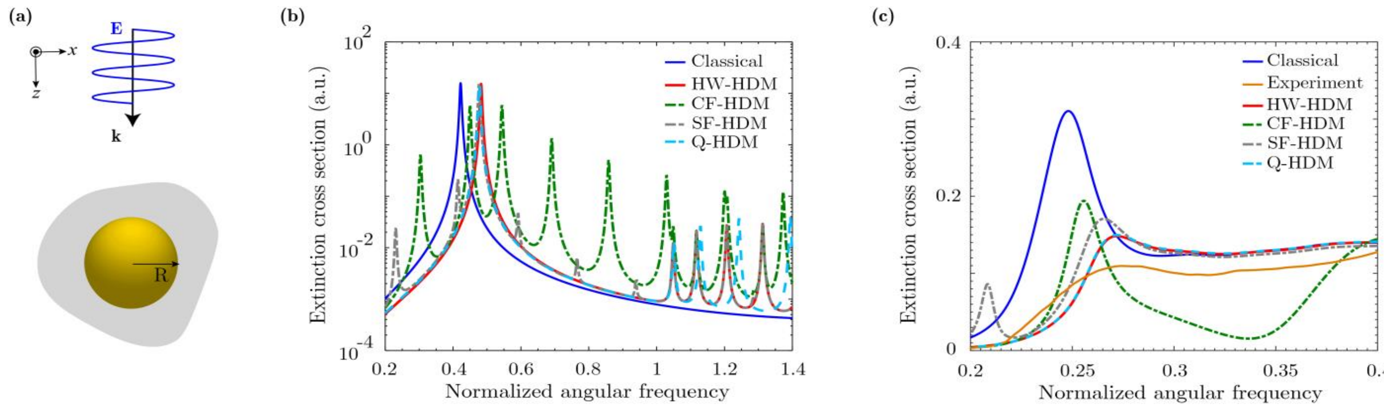

Fig. 1. Hydrodynamic analysis of a gold nanosphere: (a) the nanosphere with the radius R=1.45 nm embedded in glass and a plane wave excitation. Extinction cross section, normalized to the geometrical cross section of the sphere, is shown for (b) Drude and (c) experimentally measured material models. The results of the classical approach (no nonlocal effects) and experiments are employed as references [4].

IV. Discussion

Considering the response of free electrons, the main hydrodynamic surface plasmon resonances are blueshifted with respect to the classical one. As depicted in Fig. 1 (b), CF-HDM introduces the greatest blueshift, deviating from the other three hydrodynamic models. Moreover, above the plasma frequency, one may observe the propagating longitudinal wave modes. Four longitudinal resonances at spectral positions 1.05, 1.12, 1.21, and 1.31, may be clearly distinguished for HW-HDM. These resonances are identical to the ones of SF-HDM and are in good agreement with CF-HDM. Note that Q-HDM generates the greatest displacement of the longitudinal peaks, due to the fact that this model considers both quantum pressure and diffraction effects. Apart from the abovementioned resonances, CF-HDM and SF-HDM yield several other spurious resonances both below and above the plasma frequency, which appear more strikingly for CF-HDM. Such resonances occur since CF-HDM and SF-HDM consider, on top of the nonlocal longitudinal response (as for the other two hydrodynamic models), the nonlocal transverse response [4].

Compared with the abovementioned Drude case, the response of both free and bound electrons in the studied frequency range, described by an experimental material model [5], yields the following two differences: 1) the classical and nonlocal hydrodynamic surface plasmon resonances have been redshifted and 2) their magnitudes have been significantly attenuated, as illustrated in Fig. 1 (c). The results of HW-HDM and Q-HDM are almost the same, while the other two hydrodynamic models deviate considerably. Compared with HW-HDM and Q-HDM, SF-HDM generates a nearly identical nanoparticle's response above a spectral position of 0.3. However, it predicts a slightly lower blueshift of the surface plasmon resonance, and more surprisingly, this model reveals another distinguishable resonance peak at approximately 0.23. CF-HDM clearly introduces the greatest discrepancy in the spectrum.

Finally, the results of hydrodynamic models are compared with experiments available in literature for the studied topology. As it can be seen in Fig. 1 (c), the reported experimental results in general agree closely with HW-HDM and Q-HDM, and do not show spurious components generated by the other two hydrodynamic models.

V. Conclusion

This work performed a comparison of four nonlocal hydrodynamic models, namely HW-HDM, CF-HDM, SF-HDM, and Q-HDM. This was done by investigating the response of a metallic nanosphere with a spatially dispersive material parameter, under a plane wave excitation. First, a Drude material model was employed, considering the response of only free electrons. Second, an experimentally measured material model was employed, in order to include the response of both free and bound electrons. It was shown that different hydrodynamic models may predict the particle's response differently. CF-HDM and SF-HDM display anomalous features in the extinction spectrum, which do not show up for the other two hydrodynamic models. Additionally, the results of hydrodynamic models were compared with experimental results reported in literature, yielding a good agreement with HW-HDM and Q-HDM.

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