Surface Science 637–638 (2015) 53–57

Contents lists available at ScienceDirect

Surface Science

journal homepage: www.elsevier.com/locate/susc

Plasmon modes of spherical nanoparticles: The effects of quantum nonlocality

Afshin Moradi *

Department of Engineering Physics, Kermanshah University of Technology, Kermanshah, Iran

ARTICLE INFO

Article history:
Received 27 January 2015
Accepted 9 March 2015
Available online 18 March 2015

Keywords:
Quantum nonlocal effect
Quantum hydrodynamical model
Plasmon

ABSTRACT

We develop a new method for calculating the electrostatic surface and bulk plasmon modes of a spherical metal nanoparticle, by taking into account the quantum nonlocal effects. To describe these phenomena, we develop analytical theory based on the quantum hydrodynamical model of plasmon excitation. We derive new dispersion relation for the system and investigate its differences with previous treatments based on the standard nonlocal model.

1. Introduction

It has been realized for a long time that small metallic spheres can support surface plasma (SP) oscillations [1]. Also, in the local model [2,3] it is well-known that metallic spherical nanoparticles in vacuum, exhibit resonant absorption of light at the dipole SP frequency $\omega_1 = \omega_p/\sqrt{3}$, where ω_p, is the plasma frequency. This frequency, is just the first in a series of SP modes of the sphere, as $\omega_\ell = \omega_p\sqrt{\ell/(2\ell+1)}$ [4–6]. An important next step was made in Ref. [7], where the standard hydrodynamic (SHD) model of electron gas was applied to study the plasmons in metallic spheres. However, it was limited in applications to the nondispersive limit [8]. Then the investigation of the nonlocal effects in the response function of small metal spheres has been followed by Lushnikov and Simonov [9], using the quantum mechanical random-phase approximation (RPA) and by Ascarelli and Cini [10] with the SHD model [7,8]. Dasgupta [11] obtained a pair of matrix equations describing the SP modes dispersion for a very small spherical metal particle, by means of the quantum mechanical RPA. Furthermore, Ruppin [12,13], in the presence of the spatial dispersion and surface diffuseness, calculated the frequencies of the plasmons of small metal spheres, by employing the SHD theory. In this way, Ogale et al. [14], derived the SP modes dispersion relation for spherical metallic particles in the following two cases: (1) a sharp surface cut off in electron density and (2) a diffused electron density at the surface. Dasgupta and Fuchs [15], developed a simple semiclassical method for calculating the polarizability of a very small spherical particle by taking into account the nonlocal nature of the dielectric response. Barberan and Bausells [16], studied the collective modes of metal sphere, including bulk-type modes [17], using the SHD model. Fuchs and Claro [18], investigated the multipolar response of a small metallic sphere with use of a nonlocal dielectric function.

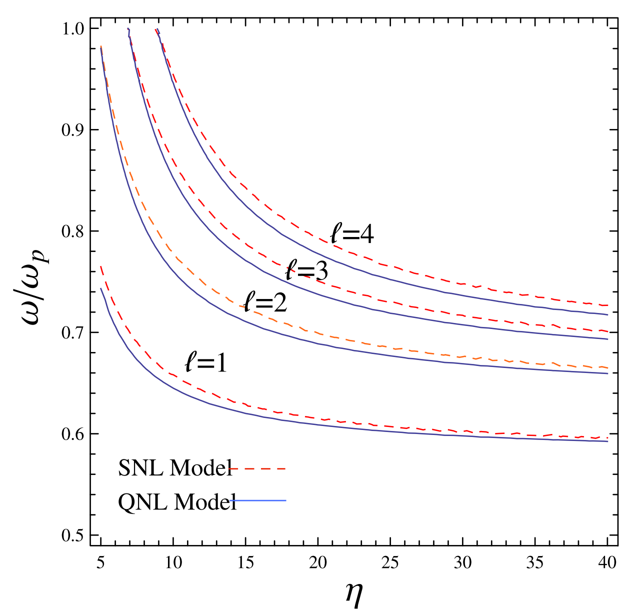

Fig. 1. Dispersion curves of surface modes ($n = 0$) in a spherical nanoparticle for different values of the parameter ℓ vs. the variable $\eta = a\omega_p/\alpha$. For the other parameters the sodium values have been employed. For $\ell = 0$ the surface mode is not possible. The results for the SNL dispersion relation are shown by the dashed red curves. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

* Corresponding author. Tel.: +98 9183312692.
E-mail address: a.moradi@kut.ac.ir.

http://dx.doi.org/10.1016/j.susc.2015.03.005
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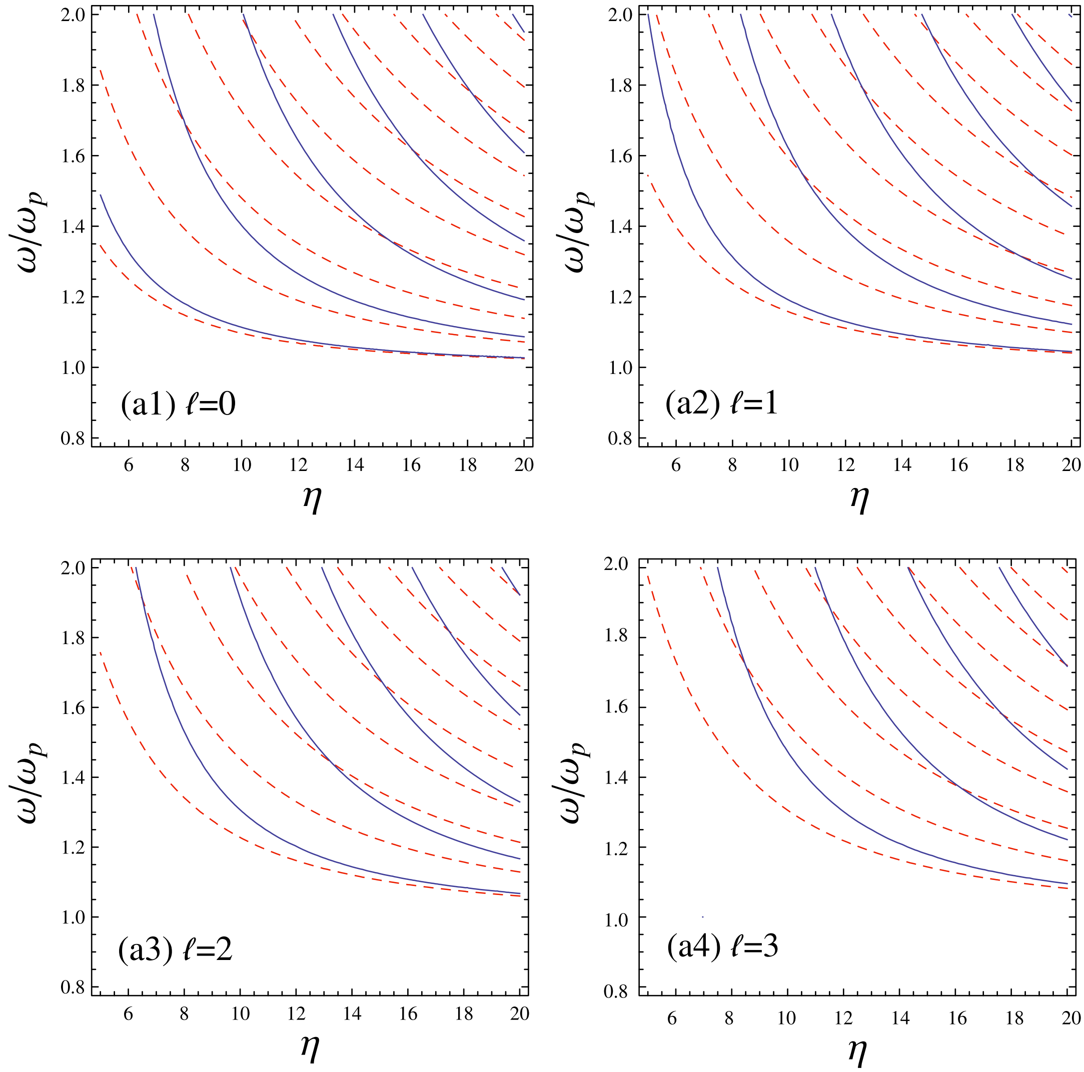

Fig. 2. Same as Fig. 1, but for bulk modes ($n \neq 0$). Panels (a1)–(a4) illustrate different values of the parameter ℓ.

However, it is well-known that the classical electronic pressure in the SHD model may not suffice for studying the nonlocal effects for electron excitations in nanometer sized structures [19–25]. Generally, the quantum effects should be considered in an electron fluid system when the de Broglie wavelength associated with the electrons is comparable to dimension of the system. Among the different theoretical models for the study of the quantum effects in the solid state plasma, the quantum hydrodynamic (QHD) [26–30] has become popular for its extension of the usual fluid model to one incorporating the quantum effects.

In the present work, following our previous paper [23–25] and motivated by the recent works given in Refs. [19–22], we study the quantum nonlocal effects on the optical properties of spherical metal nanoparticles by means of the linearized QHD theory. In this way, the frequency of surface and bulk plasmon modes of system can be determined in the non-retarded (electrostatic) approximation by solving Laplace equation with suitable additional quantum boundary conditions for the surface charge densities, according to the more recent results obtained by Miskovic et al. [22]. Let us note that the electrostatic approximation, which neglects the effects of retardation, is valid for the systems whose dimensions are much smaller than the wavelength of incident light.

The rest of this paper is presented in the following way. In Section 2, we derive main equations of our model. Numerical results are discussed in Section 3. We conclude with a summary of our results in Section 4.

2. Basic theory

In order to carry out the calculations of the quantum effects on the surface and bulk plasmon modes of a spherical metal nanoparticle of radius a bounded by vacuum, we start with the linearized QHD theory of an electron gas [22–24]. The appropriate equations; (1) the force equation, (2) Poisson equation, and (3) the continuity equation; are

$$\frac{\partial}{\partial t}\Psi(\mathbf{r},t) = -\frac{\omega_p^2}{4\pi e n_0}\Phi(\mathbf{r},t) + \frac{\alpha^2}{n_0}n(\mathbf{r},t) - \frac{\beta^2}{n_0}\left[\nabla^2 n(\mathbf{r},t)\right], \tag{1}$$

$$\nabla^2\Phi(\mathbf{r},t) = 4\pi e n(\mathbf{r},t), \tag{2}$$

$$\nabla^2\Psi(\mathbf{r},t) = \frac{1}{n_0}\frac{\partial}{\partial t}n(\mathbf{r},t), \tag{3}$$