Optics Communications 326 (2014) 105–113

Contents lists available at ScienceDirect

Optics Communications

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

Nonlinear continuum mechanical model for investigating plasmonic oscillations phenomena in nanostructured metals

Aleksey M. Serebrennikov [a,b,*]

[a] Department of General Physics, Perm National Research Polytechnic University, Komsomolsky Avenue, 29, 614990 Perm, Russia
[b] Department of Automation and Telemechanics, Perm National Research Polytechnic University, Komsomolsky Avenue, 29, 614990 Perm, Russia

ARTICLE INFO

Article history:
Received 4 January 2014
Received in revised form
2 April 2014
Accepted 7 April 2014
Available online 21 April 2014

Keywords:
Nonlinear plasmonics
Metal nanoparticles
Third-harmonic generation
Kerr nonlinearity

ABSTRACT

Here, we introduce a nonlinear continuum mechanical theoretical model of dissipative plasmonic oscillations relying on the principle of least action. The proposed theory has allowed obtaining the expression of a stress tensor for an "electron gas – ionic frame" system. In parallel, an initial boundary value problem for nonlinear integrodifferential equations constituting the model has been formulated. On the basis of a finite-difference approach the iterative solution method, algorithm and solver have been worked out. Thereby we have investigated the phenomena of harmonic multiples generation by a cluster of metal nanoparticles. Also by using these tools the estimate of the density function parameter satisfying the requirement of regular oscillations has been obtained numerically. On the ground of extensive numerical runs it was found that for a given set of parameters the system response turned out to be mainly linear, however the contributions of the closest odd harmonic multiples (third and fifth) were well resolved under quantitative analysis. This result allows the nonlinearity governable by the principal equation of motion to be associated with Kerr's type nonlinearity.

1. Introduction

In this work the theoretical (mathematical) model of plasmonic oscillations is evolved on the basis of the density function approach proposed first in [1]. The considered theory has been applied to the characterization of nonlinear plasmonic oscillations in metal nanoparticles (MNP) subjected to the illumination by an intense light. The attention of the continuum mechanics community to the phenomenon of plasmonic oscillations in metals can be explained by the obvious fact that its theoretical investigation traditionally (for more than a century) relies on the methods of classical particle mechanics [2]. At present plasmonics (an interdisciplinary research domain covering fundamental and application aspects of plasmonic behaviors) presents a rapidly growing area of optoelectronics having quite a number of transitions into the different branches of material sciences such as biophysics, colloidal chemistry, continuum mechanics, optomechatronics and many more. Its perspectives are envisioned in the areas of telecommunications, computers, medical therapy, photovoltaics, spectroscopy, etc.

The objects of interest in nonlinear plasmonics are first of all the processes ordinarily investigated in nonlinear optics such as the harmonic multiples generation, wave mixing, solitons and the control of light-by-light. As it was established, besides other capabilities the approach proposed in this work allows the alternative description of the Kerr effect as compared with the standard nonlinear optics. Let us recall that when a nonlinear system illuminated with the monochromatic electromagnetic field of frequency ω generates the scattered (secondary) field as a superposition of the fundamental (ω) and triple (3ω) frequency harmonics, it is considered to be "Kerr-responding". This effect has a simple interpretation in the framework of the theory of nonlinear polarizability [3]. Indeed, the polarizability function **P(E)** can be expanded into the power series of the electric field **E** up to third order inclusive and in this form goes to the field equations. Under the temporal waveform $\cos(\omega t)$ excitation, the field equations contain the terms $\cos^3(\omega t)$. The latter can be converted into the linear combination of $\cos(\omega t)$ and $\cos(3\omega t)$ functions by trigonometrical transforms. Thereby the field equations predict the excitation of the first and third harmonics mixture in the scattered electromagnetic spectra as a temporal first order effect. In our case the mathematical description of the Kerr effect has no common points with the theory of nonlinear polarizability; its appearance is completely governed by the construction of the energy terms of the Lagrange function used in the least action principle.

* Correspondence address: Department of General Physics, Perm National Research Polytechnic University, Komsomolsky Avenue, 29, 614990 Perm, Russia.
E-mail address: al-serebr02@yandex.ru

http://dx.doi.org/10.1016/j.optcom.2014.04.010
0030-4018/

Speaking about the application of continuum mechanical models for the description of linear and nonlinear properties of plasmonic metals it is worth to note that the nonlocal hydrodynamic Drude model based on the Navier–Stokes type equation has seemingly entered into the most widespread use [4–7]. The main area of its utilization is the studies on the plasmonic nonlocality where the equation is applied in the linearized form [4,5]. In [6,7] the contribution of the convective term into the second harmonic-generation in metals has been investigated. In these works the impacts of surface and ion core electrons have been taken into account in the framework of the theory of nonlinear polarizability [3].

In the present paper the model of nonlinear dynamics of the gas of valence electrons (metal plasma) is widened as compared with that introduced in [1]. Explicitly, axiomatics of the theory is formulated with more rigor. The dissipation function of a polynomial type has been embedded into the structure of the action functional of an "electron gas–ionic frame" system. The denotation of a stress tensor for the system experiencing plasmonic oscillations is introduced. An initial boundary value problem has been formulated for the system of integrodifferential equations of nonlinear mechanics derived by the least action principle. On the ground of a finite-difference approach the numerical method and algorithm for treating the problem have been worked out. By the examples of harmonic multiples generation in the electromagnetic spectrum the investigation of motion of an electronic continuum under different values of the density function parameter and dissipative force coefficients is fulfilled.

2. Axiomatics of the theoretical model of the metal plasma nonlinear dynamics

The principal question risen in the present (and partially in the previous [1]) work is the one touching the possibility of constructing mechanics based on miscellaneous (hybrid) axiomatics. Some positions of this mechanics rest on the axioms of standard continuum mechanics; others rely on those borrowed from quantum mechanics. As is known, the most complete method of characterization of a system from all possible ones in quantum mechanics is its description with the use of the wave function. Rigorously speaking, the latter is achievable only for isolated systems. In practice, a quantum system attracting the research interest usually presents a part of a certain larger isolated system. The characterization of such subsystems is performed by using their density matrices [8]:

$$\rho(\mathbf{r},\mathbf{r}',t)=\int_Q \Phi(\mathbf{r},\mathbf{q},t)\,\Phi^*(\mathbf{r}',\mathbf{q},t)\,d\mathbf{q}, \tag{1}$$

where $\Phi(\mathbf{r},\mathbf{q},t)$ is the many − body wave function, $\mathbf{r}$ and $\mathbf{r}'$ – the coordinates of a subsystem, $\mathbf{q}$ – all the rest of the coordinates of a larger system, and Q – the volume of a configuration space. The notations $\mathbf{r}$ and $\mathbf{q}$ besides spatial coordinates include particle spins. Also notation (1) besides the integration itself assumes the summation over spins. The density function serves as a particular case of the density matrix being its diagonal element $\rho(\mathbf{r},\mathbf{r},t)$ with the reduced dimensionality of coordinates: $\mathbf{r}\in R^3$. The description of quantum systems by means of the density function underlies the *density functional theory* [9]. This approach leads to the system of nonlinear differential (Kohn–Sham) equations which are quite close in structure to the Hartree–Fock equations. Mathematically the function $\rho(\mathbf{r},\mathbf{r},t)$ (the second argument will be neglected henceforth so that $\rho(\mathbf{r},\mathbf{r},t)=\rho(\mathbf{r},t)$) is continuous and defined everywhere in space. Thereby a commonality between the quantum mechanics and classical continuum mechanics in the characterization of matter emerges: both mechanics deal with the "continuum of density". This inference allows quantitative equalization of charge (mass) and current (momentum) density functions of both mechanics [1]:

$$\rho_{continuum}(\mathbf{r},t)=\rho_{quant}(\mathbf{r},t),\quad \mathbf{j}_{continuum}(\mathbf{r},t)=\mathbf{j}_{quant}(\mathbf{r},t),$$

where the quantum mechanical current density

$$\mathbf{j}_{quant}(\mathbf{r},t)=\frac{e}{m}\left\{\frac{i\hbar K}{2}\int_Q(\Phi\nabla\Phi^*-\Phi^*\nabla\Phi)\,d\mathbf{q}-\rho_{quant}\mathbf{A}\right\} \tag{2}$$

results from differentiation of Eq. (1) with the simultaneous use of the wave equation:

$$i\hbar\frac{\partial\Phi}{\partial t}=\sum_{k=1}^{K}\left\{\frac{(\hat{\mathbf{p}}_k-e\mathbf{A}(\mathbf{r}_k,t))^2}{2m}+e\,\psi(\mathbf{r}_k,t)+U\right\}\Phi, \tag{3}$$

where $\hat{\mathbf{p}}_k=-i\hbar\nabla_k$ is the momentum operator, U – the total Coulomb potential, and $\mathbf{A}(\mathbf{r},t)$ and $\psi(\mathbf{r},t)$ – the vector and scalar electromagnetic potentials. (Rigorously speaking Eqs. (2) and (3) relate to the quantum system composed from $\mathbf{K}$ identical fermions interacting with nuclei being resting in the framework of adiabatic approximation. In this sense Eqs. (2) and (3) can be applied for studying a complete electron–nucleus system). The discreteness of energy spectrum is the fundamental concept of quantum mechanics, however, for the objects having three linear dimensions of order of tens of nanometers (composed from $10^6/10^7$ atoms) and larger, the energy spectrum can be conceived of as a quasi-continuous. As a consequence, the energy quantities of the two mechanics can also be equalized for such objects. (Albeit for some physical phenomena such as high-temperature superconductivity this statement may not be valid.)

Let now axiomatically introduce the density function as $\rho(\mathbf{u},\mathbf{u}\nabla,t)$, where $\mathbf{u}(\mathbf{r},t)\in(C^2(R^4))^3$ is a certain vector field defined everywhere. The density defined on the element $\mathbf{u}(\mathbf{r},t)$ becomes at this point a functional. Let the field $\mathbf{u}(\mathbf{r},t)$ be referred to as a state function (or simply a state). Thus the state unambiguously determines the distribution of the density of matter in a whole space. Immediately the functional $\rho(\mathbf{u},\mathbf{u}\nabla,t)$ turns out to be defined and continuous everywhere in R^3. Thereby the state $\mathbf{u}(\mathbf{r},t)$ defined by this method serves as a certain generalization of the standard continuum mechanical displacement function. The difference between them is due to the infinite spanning of the whole space by the density function defined by the procedure specified. Under assumption that the continuum occupies the finite piece of space (this admission has been made in [1]) one can refer to its initial and current configuration as Ω_0 and Ω_1, thus the state can be conceived of as a continuous mapping $\mathbf{u}(\mathbf{r},t):\Omega_0\to\Omega_1$ (that allows referring to it as a displacement function). With this mapping besides the translation movement of the continuum an alteration of its shape and volume arises. Consequently, one may speak about the continuum deformation and the dependence of its density upon strains. As a measure of strains the tensor field $\mathbf{u}\nabla=u_{i,j}\,\mathbf{e}_i\mathbf{e}_j$ has been chosen in [1]. Thereby the appearance of $\mathbf{u}\nabla$ in the list of arguments of ρ can be explained. In case of the density defined in the whole R^3 and vanishing only at infinity, a methodological difficulty arises in the formulation of initial and current configurations of the continuum, because the movement of density does not now lead to the volume and shape alteration of the "whole" R^3. Therefore for this moment a postulation of the dependence of ρ on $\mathbf{u}\nabla$ has the formal character and needs finding other motivations (we will come back to this point in Section 3 of this paper). In view of the above, the field $\mathbf{u}(\mathbf{r},t)$ becomes devoid of kinematical sense being currently an internal characteristic of state of the mechanical system. The knowledge of $\mathbf{u}$ does not now allow analyzing the system motion by means of the "visual image" of this function. Thus a motivation arises to formulate the measures of motion required to maintain this goal.