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Scattering of electromagnetic pulses by metal nanospheres in the vicinity of a Fano-like resonance

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ABSTRACT

In the work, radiation scattering by metal nanospheres in a dielectric matrix in case of ultrashort and long electromagnetic pulses is studied theoretically. Spectral efficiencies of backward and forward scattering by silver nanospheres in glass are calculated with the use of experimental data on the dielectric permittivity of silver. The presence of Fano-like resonances in spectral dependences of scattering efficiency caused by interference of dipole and quadrupole scatterings is shown. Backward and forward scattering of ultrashort pulses is calculated and analyzed. The obtained dependences of the total probability of scattering (during all time of the action of a pulse) on pulse duration demonstrate an essential distinction between an ultrashort case and a long pulse limit.

An autoionization resonance in inelastic scattering of electrons by atoms was studied in the classical work [1], in which, on the basis of the consistent quantum-mechanical theory, well-known expression for a Fano profile was obtained. Autoionization resonances with a Fano profile in excitation of a helium atom by synchrotron radiation were experimentally recorded half a century ago in the work [2].

Recently, in connection with rapid progress in technology of generation of ultrashort electromagnetic pulses (USP) [3], the study of USP interaction with a substance has become topical, in particular, for spectral ranges corresponding to autoionization resonances. For example, in the paper [4] excitation of helium atom under the action of an attosecond UV pulse and of a time-shifted few-cycle IR pulse was analyzed within the framework of a one-dimensional model. In the work [5], autoionization of argon atoms by an isolated attosecond pulse in the presence of an intensive few-cycle IR laser pulse was studied by the method of transient absorption spectroscopy.

Due to development of nanotechnologies, the area of study of Fano resonances has expanded and now covers a wide range of phenomena in nanostructures and nanoparticles such as metal nanospheres, plasmon nanocavities, atomic clusters, photon crystals, etc. [6].

In the work [7] it is shown that Fano resonances arise in the spectral-angular cross-section of radiation scattering by metal nanoparticles. Forward and backward scattering of radiation was considered. The calculation was carried out according to the Mie theory. For dielectric permittivity the Drude formula was used. The presence of resonances with a Fano profile for metals with low dissipation was shown. A physical cause of appearance of an asymmetric resonance structure in this case is interference of contributions of different multipole nature to the scattering process.

The analysis shows that for real metals used in numerous applications (silver, gold, copper) the Drude formula in the frequency range of Fano-like resonances ($\hbar\omega > 2$ eV) poorly describes the dielectric permittivity because of the considerable contribution of interband electronic transitions.

The present work is dedicated to the study of scattering (forward and backward) both of long and short pulses by metal nanospheres in the region of a resonance caused by interference of dipole and quadrupole contributions to scattering.

Let us expand the electric field of a USP to monochromatic components, then for the scattering probability during all time of the action of a pulse the following expression can be obtained [8,9]:

$$W_{sc} = \int_0^\infty \sigma_{sc}(\omega') \frac{dN_{ph}}{ds\, d\omega'} d\omega', \qquad (1)$$

where $\sigma_{sc}(\omega')$ is the spectral cross-section of scattering,

$$\frac{dN_{ph}}{ds\, d\omega'} = \frac{c}{(2\pi)^2} \frac{|E(\omega')|^2}{\hbar\omega'} \qquad (2)$$

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is the number of photons at a frequency ω' that passed through an area element ds during USP scattering by a target. Substituting the formula (2) in the expression (1), we find:

$$W_{sc} = \frac{c}{(2\pi)^2} \int_0^\infty \sigma_{sc}(\omega') \frac{|E(\omega')|^2}{\hbar\omega'} d\omega' \tag{3}$$

In the case under consideration for radiation scattering by a nanosphere, the process cross-section can be represented as

$$\sigma_{sc} = \pi r_s^2 Q_{sc} \tag{4}$$

r_s is the nanosphere radius, Q_{sc} is the radiation scattering efficiency.

The expression for the probability of (radar) backward and forward scattering of an ultrashort pulse by a metal nanosphere follows from the formulas (3)–(4):

$$W_{RBS(FS)} = \frac{c}{4\pi^2} \int_0^\infty \pi r_s^2 Q_{RBS(FS)}(\omega') \frac{|E(\omega')|^2}{\hbar\omega'} d\omega', \tag{5}$$

here

$$Q_{RBS} = \frac{1}{x^2} \left| \sum_{n=1}^\infty (2n+1)(-1)^n [a_n - b_n] \right|^2, \tag{6}$$

$$Q_{FS} = \frac{1}{x^2} \left| \sum_{n=1}^\infty (2n+1)[a_n + b_n] \right|^2 \tag{7}$$

are the efficiencies of backward and forward scattering of monochromatic radiation [7], $x = \sqrt{\varepsilon_d} \cdot r_s\omega/c$ is the dielectric permittivity of a matrix, in which nanoparticles are placed. The scattering amplitudes a_n, b_n are given by the known formulas of the Mie theory [10]. The functions a_n, b_n depend on two parameters: x and $m = \sqrt{\varepsilon_s(\omega)/\varepsilon_d}$, where $\varepsilon_s(\omega)$ is the dielectric permittivity of a nanosphere material.

The expression for the scattering efficiency integrated with respect to the scattering angle is

$$Q_{\rm int} = \frac{2}{x^2} \sum_{n=1}^\infty (2n+1)\left[|a_n|^2 + |b_n|^2\right]. \tag{8}$$

Let us calculate the probability of backward and forward scattering of an ultrashort pulse of a corrected Gaussian shape proposed in the work [11]. The Fourier transform of the strength of an electric field in such a pulse is given by the expression

$$E_{cor}(\omega', \omega) = iE_0\tau \sqrt{\frac{\pi}{2}} \frac{\omega'^2\tau^2}{1+\omega^2\tau^2} \times \left\{e^{-i\varphi-(\omega-\omega')^2\tau^2/2} - e^{i\varphi-(\omega+\omega')^2\tau^2/2}\right\}, \tag{9}$$

where ω', ω are the current and carrier frequencies, E_0, τ are the amplitude of the electric field strength and the pulse duration, φ is the carrier phase with respect to the envelope. It should be noted that a corrected Gaussian pulse does not contain a constant component of the strength $E_{cor}(\omega' = 0, \omega) = 0$ in contrast to a traditional Gaussian pulse and at the same time allows a limiting transition to the region of long pulses.

As a target, we will consider a silver nanosphere placed in a glass matrix. The results of calculation of the efficiency of backward and forward scattering of monochromatic radiation according to the formulas (6), (7) for a nanosphere of radius 40 nm are presented in Fig. 1. Shown in the same figure is the scattering efficiency integrated with respect to the angle (8), normalized to a corresponding factor. The dielectric permittivity of a nanosphere material $\varepsilon_s(\omega)$ was calculated with the use of experimental data of the work [12].

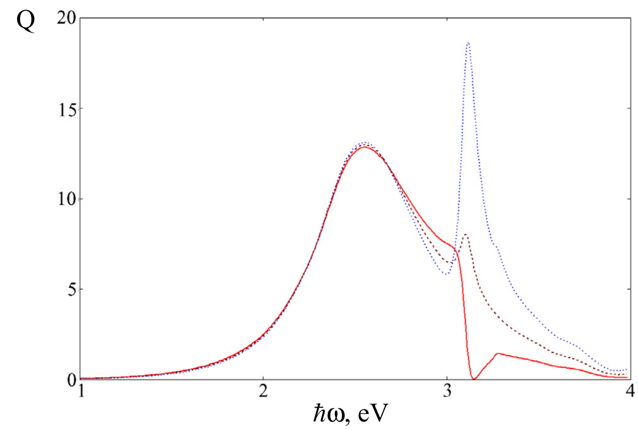

Fig. 1. The efficiencies of backward (solid curve) and forward (dotted curve) scattering of radiation and the scattering efficiency integrated with respect to the angle (dashed curve) for a silver nanosphere of radius 40 nm in glass.

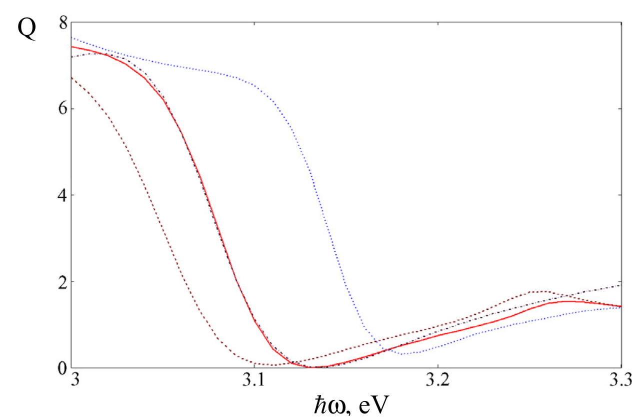

Fig. 2. The interference minimum in backward scattering in the region of a quadrupole resonance for silver spheres of different radii in glass: solid curve – $r_s = 42$ nm, dotted curve – $r_s = 35$ nm, dashed curve – $r_s = 45$ nm, dash-and-dot curve – fitting by a Fano profile for $r_s = 42$ nm.

From the given figure it follows that in the region of a dipole maximum the presented efficiencies practically coincide with each other. At photon energy about 3.2 eV, the backward scattering efficiency has a deep minimum due to destructive interference of dipole and quadrupole contributions to the process. Practically at the same frequency the forward scattering efficiency has a strong maximum due to constructive interference of dipole and quadrupole scattering.

Presented in Fig. 2 are the spectral dependences of the efficiency of backward scattering of radiation by silver nanospheres of different radii in the region of a quadrupole resonance, where there is an interference minimum. It is seen that with increasing nanosphere radius the position of a minimum is shifted to the low-frequency region, and there is an optimum value of radius r_s^{opt}, for which a minimum is the deepest.

Besides, fitting of the scattering efficiency with the use of a Fano profile (10) is given for a nanosphere of radius $r_s = 42$ nm. The formula for fitting looks like:

$$F(\omega) = A \frac{[(\omega-\omega_0)/\Gamma + q/2]^2}{1/4 + [(\omega-\omega_0)/\Gamma]^2} + B \tag{10}$$

It should be noted that for $A = 1$ and $B = 0$ the expression (10) represents a classical Fano profile. The following parameters were used for fitting: $\hbar\omega_0 = 3.077$ eV, $\hbar\Gamma = 0.12$ eV, $q = -0.97$, $A = 3.74$, $B = 0$.