IRAMP
6(1), June 2015, pp. 1-6

ISSN: 2229-3159

Absorption of ultrashort electromagnetic pulses by metal clusters

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ABSTRACT: The probability of absorption of an ultrashort pulse by metal clusters in the vicinity of a plasmon resonance during the action of a pulse is calculated and analyzed. A case of pulses with a carrier frequency and electromagnetic bursts (wavelets) without a carrier frequency is considered. The features of the time dependence of the process probability that are characteristic of short pulse durations are revealed.

1. INTRODUCTION

The development of the technique of generation of ultrashort electromagnetic pulses (USP) necessitates the study of features of their interaction with various targets [1]. The present paper is dedicated to the theoretical study of USP absorption by metal clusters. A feature of such clusters is that their valence electrons are not localized in space and can be considered as being in the field of positively charged ions. For simplification of consideration of such structures, various models are proposed, for example, a so-called "jelly model" [2] that implies that ions are distributed uniformly and spherically symmetrically in some volume of space, and in their field sharing (valence) electrons move. Under the action of external radiation at a specific frequency, valence electrons make collective oscillations near the cluster surface, that is, surface plasmons are excited [3], the characteristic frequency of which (in energy units) is several electron-volts. It should be noted that there are also other mechanisms of generation of plasmons in atomic clusters. For example, in xenon clusters plasmon excitations are formed as a result of collective motion of electrons of the 4d shell [4], in this case the plasmon energy is about 100 eV.

The radius of a metal cluster depends on the number of atoms in the latter (N) and in a typical case is about 1 nm and less (for $N \sim$ 10-100). Thereby metal clusters differ from metal nanospheres, the characteristic radius of which is usually from 10 to 50 nm. Since the number of atoms in metal nanospheres is sufficiently great, and there is a sharp boundary, the Mie theory can be used to describe their interaction with an electromagnetic field of the optical range. USP absorption by metal nanospheres put in a dielectric medium was studied in the work [5]. In case of metal clusters with a small number of atoms, when there is no sharp boundary, the calculation of absorption of electromagnetic radiation by these clusters requires another approach.

In this work, the features of USP absorption by metal clusters of small radius ($R < 1$ *nm*) in the vicinity of a plasmon resonance during the action of an electromagnetic field are studied within the framework of the first order of the perturbation theory.

2. METHOD OF CALCULATION

The probability of excitation of an atom (ion) under the action of an ultrashort pulse in the first order of the perturbation theory for interaction with an electromagnetic field is given by the following expression [6]:

$$W = \frac{c}{4\pi^2}\int_0^\infty \sigma(\omega')\frac{|E(\omega')|^2}{\hbar\omega'}d\omega', \tag{1}$$

where c is the velocity of light, ω' is the current frequency, $\sigma(\omega')$ is the absorption cross-section, $E(\omega')$ is the Fourier transform of the strength of the electric field in an electromagnetic pulse. It is obvious that the expression (1) makes sense in the framework of applicability of the perturbation theory, that is, at $W < 1$.

As follows from the optical theorem [2], the photoabsorption cross-section is expressed in terms of the dynamic polarizability $\alpha(\omega)$ according to the formula:

$$\sigma_{abs}(\omega) = \frac{4\pi\omega}{c}\operatorname{Im}\alpha(\omega). \tag{2}$$

The dynamic polarizability of a metal cluster in the resonant plasmon approximation is [7, 8]

$$\alpha(\omega) = R^3\frac{\omega_p^2}{\omega_p^2 - \omega^2 - i\omega\ \Gamma}, \tag{3}$$

where ω_p is the resonance frequency of a surface plasmon, Γ is the width of the resonance curve.

The radius of a metal cluster R in the jelly model is [2]

$$R(N) = r_{ws}N^{1/3}, \tag{4}$$

where N is the number of atoms in a cluster, r_{ws} is the size of a Wigner-Seitz cell of a metal.

The plasmon resonance frequency is given by the following expression:

$$\omega_p(N) = \sqrt{\frac{N}{\chi\ R^3(N)}}, \tag{5}$$

where χ is the factor taking into account the shift of a plasmon resonance that is caused by the screening action of ions in a metal cluster. When substituting (4) in (5), we see that in the approximation under consideration the plasmon resonance frequency does not depend on the number of atoms in a cluster:

$$\omega_p = \frac{1}{\sqrt{\chi\ r_{ws}^3}}. \tag{6}$$

The formulas (5) and (6) are written in the atomic system of units, in which $m_e = e = \hbar = 1$. Using the equations (2)-(6), we find the resultant expression for the cross-section of photoabsorption in the vicinity of a plasmon resonance:

$$\sigma_{abs}(\omega) = \frac{4\pi\ R^3}{c}\frac{\omega_p^2\omega^2\Gamma}{\left(\omega_p^2 - \omega^2\right)^2 + \omega^2\ \Gamma^2}. \tag{7}$$

Substituting (7) in (1), we obtain the formula for the total probability of USP photoabsorption by a metal cluster:

$$W = \frac{R^3\omega_p^2(R)\ \Gamma}{\pi\hbar}\int_0^\infty \frac{|E(\omega')|^2\ \omega'}{\left(\omega_p^2(R) - \omega'^2\right)^2 + \omega'^2\ \Gamma^2}d\omega'. \tag{8}$$

As ultrashort pulses acting on a metal cluster, we will consider a corrected Gaussian pulse as well as sine and cosine wavelet pulses without a carrier frequency.

The squared absolute value of the Fourier transform of a corrected Gaussian pulse looks like [9, 10]

$$\left|E_{cor}(\omega')\right|^2 \cong E_0^2\tau^2\,\frac{\pi}{2}\left[\frac{\omega'^2\tau^2}{1+\omega^2\tau^2}\right]^2\exp(-(\omega-\omega')^2\tau^2), \tag{9}$$

where ω' is the current frequency, E_0 is the amplitude of the electromagnetic field strength, τ is the pulse duration, ω is the carrier frequency.

The Fourier transforms of sine and cosine wavelets have respectively the following expressions:

$$E_s(\omega) = 2i\sqrt[4]{\pi}\omega\tau^2 E_0\exp\left(-\omega^2\tau^2/2\right), \tag{10}$$

$$E_c(\omega) = 2\sqrt{2/3}\;\;\sqrt[4]{\pi}\omega^2\tau^3 E_0\exp\left(-\omega^2\tau^2/2\right). \tag{11}$$

A feature of these wavelet pulses is the absence of carrier frequency, so their spectrum depends only on the USP duration.

Hereafter for representation of results of calculations we will use the normalized probability of photoabsorption:

$$\tilde{W} = \frac{W}{E_0^2}. \tag{12}$$

3. DISCUSSION OF RESULTS

The calculations were carried out for a Na_8 cluster, for which the resonance frequency of a surface plasmon ω_p = 2.48 eV, the resonance width $\Gamma \sim 0.1\,\omega_p$ [11], the size of a Wigner-Seitz cell r_{ws} = 3.93 a.u., χ = 1.98, R = 0.4 nm. Let us introduce the parameter of the relative detuning of the carrier frequency of USP from the plasmon frequency:

$$\delta = \frac{\left|\omega-\omega_p\right|}{\omega_p}\cdot 100\%.$$

Given below are plots with results of calculations for the normalized probability of USP absorption (12) as functions of the pulse duration and the carrier frequency.

Presented in Fig. 1 are the dependences of the normalized probability of photoabsorption of an USP of a corrected Gaussian shape on the pulse duration at different carrier frequencies of radiation. It is seen that these dependences are of nonlinear character at electromagnetic pulse durations of several femtoseconds. At longer durations the dependences go to the linear mode, which corresponds to the case of long pulses: $\omega\tau >> 1$.

From Fig. 1 it is also seen that at $\omega = \omega_p$ the dependence of the normalized probability increases quadratically (the dotted curve; to improve mapping, the scale along the ordinate axis for this curve is reduced 5 times) and at $\tau > 2$ fs goes to the linear mode. At small frequency detunings δ from the plasmon resonance frequency ω_p the slope of the curve decreases (the dash-and-dot curve, $\delta \approx 6\%$). When the detuning δ increases, an inflection appears in the solid curve in the range of USP durations 1 fs < τ < 2 fs ($\delta \approx 29\%$), where upon the curve also goes to the linear mode. In case of further increase in detuning, a characteristic maximum appears in the curve (the short dashes, $\tau \sim 1$fs; $\delta \approx 36\%$), after which going to the linear mode takes place as well.

Presented in Fig. 2 is the spectral dependence of the normalized probability of photoabsorption of an USP of a corrected Gaussian shape at different pulse durations τ.

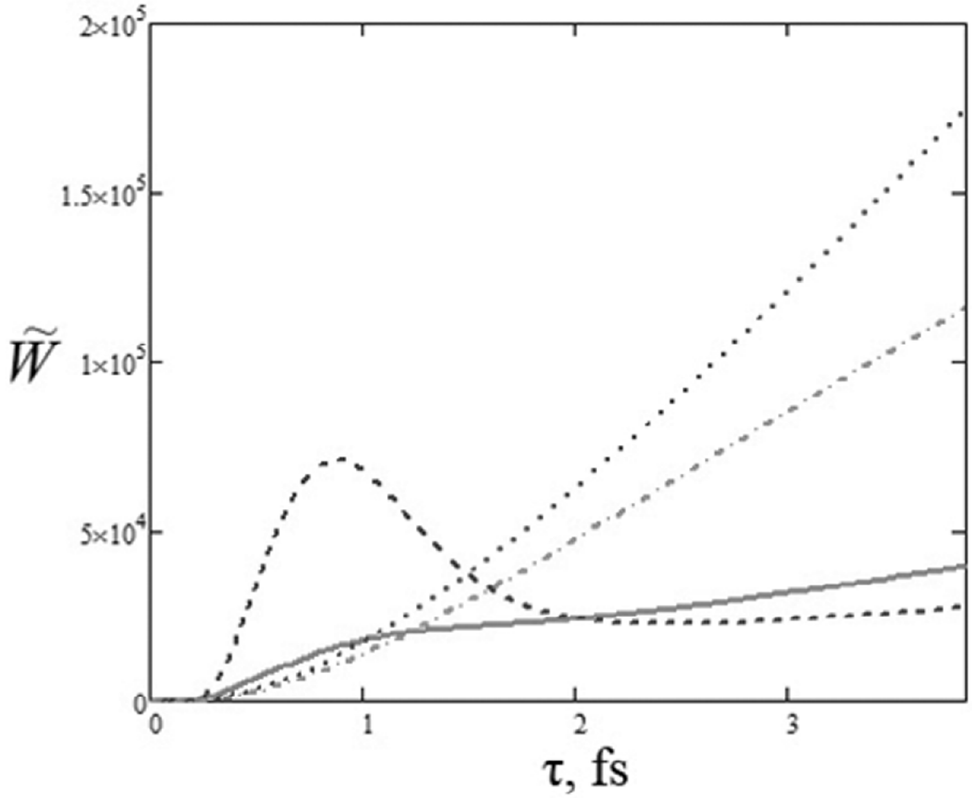

Figure 1: The dependence of the normalized probability of USP photoabsorption by a Na_8 metal cluster on the pulse duration at different frequencies of radiation: short dashes -ω = 1.63 eV, solid curve -ω = 3.26 eV, dotted curve -$\omega = \omega_p$ = 2.53 eV, dash-and-dot curve -ω = 2.68 eV. The values along the ordinate axis for the dotted and dash-and-dot curves are reduced 5 times.

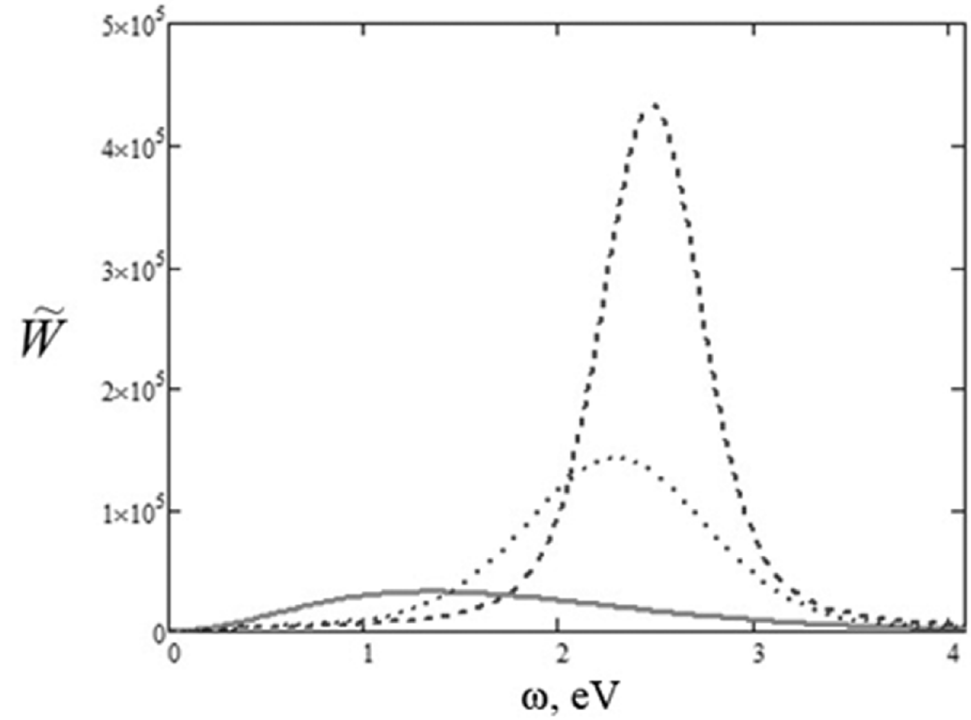

Figure 2: The spectral dependence of the normalized probability of USP photoabsorption by a Na_8 metal cluster at different USP durations: solid curve - τ = 0.48 fs, dotted curve - τ = 1.2 fs, short dashes - τ = 2.4 fs.

The dependences look like curves with a pronounced maximum, the peak value of which decreases with decreasing τ, and the peak is shifted to the low-frequency region.

Presented in Fig. 3 are the dependences of the normalized probability of USP photoabsorption (12) on the pulse duration for the case of sine and cosine wavelets without a carrier frequency (10-11). These dependences are curves with a maximum. The curve for a cosine wavelet has a larger peak value at the maximum than in case of a sine wavelet. The peak of the cosine wavelet curve is shifted to the region of long pulse durations τ.

As seen from Fig. 3, for wavelet pulses with increasing δ the dependence of the normalized probability of photoabsorption tends to zero. This is explained by the fact that with increasing δ the central frequency of the Fourier transform of a wavelet pulse is shifted to the region of lower values, which results in a decrease in the overlap of wavelet pulse spectra (10-11) and the photoabsorption cross-section (7). As a result, the value of the integral (8) defining the probability of USP absorption by a cluster in the vicinity of a plasmon resonance decreases.

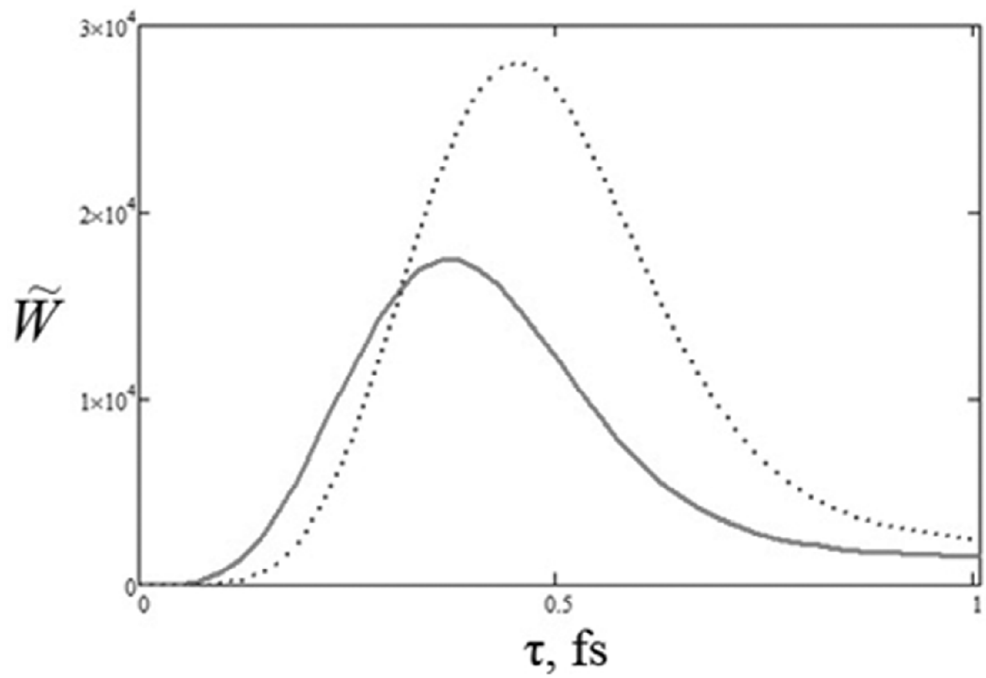

Figure 3: The dependence of the normalized total probability of USP photoabsorption by a Na_8 metal cluster on the pulse duration: solid curve-sine wavelet, dotted curve – cosine wavelet.

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