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XUV AND X-RAY ELASTIC SCATTERING OF ATTOSECOND ELECTROMAGNETIC PULSES ON ATOMS

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Abstract

Elastic scattering of electromagnetic pulses on atoms in XUV and soft X-ray ranges is considered for ultra-short pulses. The inclusion of the retardation term, non-dipole interaction and an efficient scattering tensor approximation allowed studying the scattering probability in dependence of the pulse duration for different carrier frequencies. Numerical calculations carried out for Mg, Al and Fe atoms demonstrate that the scattering probability is a highly non-linear function of the pulse duration and has extrema for pulse carrier frequencies in the vicinity of the resonance-like features of the polarization charge spectrum. Closed expressions for the non-dipole correction and the angular dependence of the scattered radiation are obtained.

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I. Introduction

The experimental achievements to generate ultra-short electromagnetic pulses (USP) with duration down to tens of attoseconds [1] have opened up new fields in modern science and technology known as attosecond physics [2] and attosecond photonics [3-4]. These short light pulses in XUV and soft X-ray range are generated with the help of optical laser produced plasmas. Special techniques based on high harmonics generation [4] allowed generation of secondary pulses with duration down to 67 as and, as shown in [5] super-continuous spectra in XUV region produced by double optical gating of high-harmonics provides the possibility to generate pulses as short as 16 as, a pulse duration being already smaller than the atomic time unit (24 as).

The current development of XFEL radiation sources demonstrated USP in broader frequency range and femtosecond durations [6]. There are also perspectives for further pulse shortening at XFEL's to sub-femtosecond of even attosecond durations: in [7] a new method was proposed to generate isolated monocycle X-ray pulses in free electron lasers with central wavelength 8.6 nm (144 eV) and pulse duration of about 50 attoseconds. Moreover, at SLAC, sub-femtosecond XFEL pulses are going to be implemented for standard users.

The theory of USP–matter interactions is challenging [8,9]. A number of works are devoted to the probability considerations of cross sections for photo processes in the field of USP [10-14]. Also USP scattering on different targets plays an important role (apart from the fundamental importance) because it may be used as a new tool for the investigation of various objects including nanoparticles [14], solid-state, complex molecules. Also bio-medical applications have been proposed [15].

One of the first works devoted to USP-scattering is the paper of V.I. Matveev [16] where the reemission spectra of USP scattering on H-like and He-like ions were studied theoretically in the framework of the sudden perturbation approximation [17]. This approximation assumes that the pulse duration is much smaller than all characteristic times of the target electron motion (sub-attosecond region). In frequency domain this means the fulfillment of the so-called high-frequency approximation that corresponds to hard X-ray photons even for relatively light atoms. In the framework of the sudden perturbation approximation USP reemission on different targets was then investigated in a number of subsequent papers (see [18] and references therein).

The dependence of the total scattering probability upon USP duration is an important characteristic feature that distinguishes the electromagnetic processes of ultra-short pulses from those of long pulses. For the latter case the probability dependence on pulse duration is linear as it follows from standard quantum mechanical treatment within the framework of the first order perturbation theory. Thus non-linearity and even resonance like features in the scattering

probability in dependence on the pulse duration is the key point in USP interaction with matter and has to be investigated in the first place. The present paper is dedicated to this question and numerical applications are carried out for the elastic (without target excitation) scattering of USP on atoms in XUV and soft X-ray range were the sudden perturbation approximation is obviously not applicable.

II. General methods of calculation: scattering tensor, non-dipole interaction and polarization charge

Let us consider the scattering of USP on atoms without the change of atomic states (elastic scattering). Including the retardation term for the electric field strength in USP we use the following expression:

$$\mathbf{E}(t,\mathbf{r}) = \mathbf{e} E_0 \, g(t - \mathbf{n}\mathbf{r}/c) \tag{1}$$

here E_0 is the field amplitude, $\mathbf{e}$ is the unit polarization vector, c is the speed of light, $\mathbf{n}$ is unit vector in the direction of the pulse propagation and $g(t_r)$ is the shape function of the incident pulse. Note, that due to the retardation term the electric field of eq. (1) satisfies automatically the wave equation (and describes a plane wave).

In the framework of the second order of perturbation theory the amplitude of spontaneous elastic scattering of radiation on atoms in state $|i\rangle$ has the form (all formulas below are in atomic units):

$$M_{sc} = \langle i|\hat{V}_f|G|\hat{V}_i|i\rangle + \langle i|\hat{V}_i|G|\hat{V}_f|i\rangle, \tag{2}$$

here $\hat{V}_{f,i}$ are perturbation operators corresponding to the interaction with the incident field (subscript i) and the scattered one (subscript f), G is atomic Green function. We consider below the scattering of USP into monochromatic plane waves with the frequency ω', polarization vector $\mathbf{e}'$ and unit vector $\mathbf{n}'$ which defines the scattering direction. From eq. (2) the following expression can be obtained:

$$M_{sc} = \sqrt{2\pi\omega'} \left(\mathbf{e}\mathbf{e}'^*\right) c_a(\mathbf{k}',\mathbf{k}) E_0 f(\omega'), \tag{3}$$

here $c_a(\mathbf{k}',\mathbf{k})$ is the atomic scattering tensor which is transformed into a scalar because of the spherical symmetry of the target, $\mathbf{k}' = (\omega'/c)\mathbf{n}'$ is the wave vector of the scattered radiation and

$\mathbf{k} = (\omega'/c)\mathbf{n}$ is the wave vector of the monochromatic component of incident USP, $f(\omega')$ is Fourier transform of the USP shape function $g(t_r)$ with respect to retarded time $t_r = t - \mathbf{n}\mathbf{r}/c$.

The quantum mechanical scattering probability is expressed via the scattering amplitude in the following way:

$$dW_{sc} = |M_{sc}|^2 d\Sigma', \tag{4}$$

here $d\Sigma' = (\omega'^2/8\pi^3 c^3) d\omega' d\Omega'$ is an element of statistical weight corresponding to the scattered radiation, $d\Omega'$ is a differential of solid angle in the direction of the scattering. As will be shown below the scattering tensor can be approximated in terms of the dynamical dipole polarizability $\alpha(\omega')$ and the normalized atomic form factor $\widetilde{F}_a$:

$$c_a(\mathbf{k}',\mathbf{k}) \approx \alpha(\omega')\widetilde{F}_a(|\mathbf{k}' - \mathbf{k}|) \tag{5}$$

Eq. (5) is a very powerful und useful non-dipole approximation that will be further illuminated below (Fig. 1). The normalized atomic form-factor is given by

$$\widetilde{F}_a(q) = \frac{1}{Z_a} \int_0^\infty \rho_a(r) \frac{\sin(qr)}{qr} dr \tag{6}$$

where $\rho_a(r)$ is radial electronic density distribution and Z_a is total number of electrons. It is convenient to introduce the polarization charge of an atom instead of polarizability according to the definition

$$Z_{pol}(\omega) = \omega^2 |\alpha(\omega)| \tag{7}$$

because in the high-frequency limit $Z_{pol}(\omega) \to Z_a$. The polarization charge is directly linked to the cross section for the radiation scattering in dipole approximation: $\sigma_{sc}(\omega) \propto Z_{pol}^2(\omega)$. Besides these relations, the polarization charge defines the spectrum of the so-called Polarization Bremsstrahlung (PB) in Bethe-Born approximation [19, 20]. This circumstance is connected with the interpretation of PB as a scattering of virtual photons from the projectile own field into real ones on the target electrons.

The polarization charge can be expressed in terms of dimensionless atomic scattering factors $f_{1,2}$:

$$Z_{pol}(\omega) = \sqrt{f_1^2(\omega) + f_2^2(\omega)}. \tag{8}$$

Eq. (8) provides a method of calculation when using atomic scattering factors that are defined and tabulated in [21] for all neutral atoms in a broad spectral range. Physically, the scattering factors $f_{1,2}$ are connected with the real and imaginary parts of the dipole dynamical polarizability of an atom, namely, $f_1=\omega^2\,\mathrm{Re}[\alpha(\omega)]$, $f_2=\omega^2\,\mathrm{Im}[\alpha(\omega)]$.

Relation (5) is exact in both limits: low-frequency and high-frequency. These limits can be obtained using the definition of the scattering tensor neglecting the eigen-frequencies in denominators and employing the completeness of the set of atomic eigenfunctions. In the low-frequency case the normalized form-factor is F=1 (dipole approximation). Therefore the scattering tensor is equal to dipole dynamic polarizability. Concerning the high-frequency case

$$c_a(\mathbf{k}',\mathbf{k}) \approx -\frac{N_e\,e^2}{m\,\omega^2}\,\widetilde{F}_a\big(|\mathbf{k}'-\mathbf{k}|\big)\ \text{where}\ \alpha\big(\omega \gg I_p\big) = -\frac{N_e\,e^2}{m\,\omega^2},$$ (I_p is the ionization potential). This

yields again Eq. (5).

To study the validity of the scattering tensor approximation (5) in the intermediate frequency domain let us compare the angle-dependent scattering factor f_1 as calculated in the framework of approximation (5) - $f_1(\omega,\theta) = f_1(\omega)\widetilde{F}_a\big(2\sin(\theta/2)(\omega/c)\big)$ and the data for $f_1(\omega,\theta)$ [21]. Figure 1 shows the results of this comparison for Al atom where the form-factor $\widetilde{F}_a(q)$ is calculated using the Hartree-Fock radial electronic density distribution with relativistic corrections, the dipole polarizability is calculated using the *dipole* scattering factors of [21]. Fig. 1 demonstrates that the multiplicative approximation for the non-dipole scattering tensor of eq. (5) is very efficient as it follows from the comparison of the non-dipole scattering factor from [21] and the non-dipole scattering factor calculated with the help of eqs. (5-8).
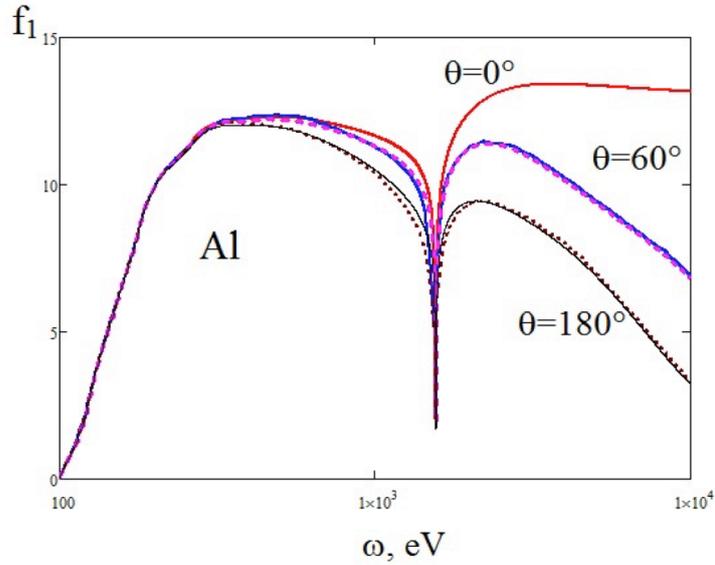

Figure 1: *Angular dependent non-dipole scattering factor f₁ for Al calculated with the scattering tensor approximation of eq. (5) (dotted blue and dotted-dashed purple curve) in comparison with the data from [21] (solid red, dashed brown and thin solid black curves).*

Naturally, the results of the two above-mentioned approaches are the same for θ=0° and we show only one curve corresponding to the scattering angle θ=0° in Fig.1. The curves for θ=60° and θ=180° demonstrate very good agreement (linear scale) between the scattering tensor approximation of eq. (5) and the reference data from [21]. Therefore, the scattering approximation (5) is identified as a very powerful one as it is exact in both limits (low and high frequency) and provides very good agreement for intermediate frequencies.

From eqs. (3)-(7) we obtain after averaging over the polarization directions an expression for the double differential probability of USP scattering on an atom during the total time of pulse action:

$$\frac{dW}{d\Omega' d\omega'} = \frac{1+\cos^2\theta}{8\pi^2 c^3 \omega'} Z_{pol}^2(\omega') E_0^2 |f(\omega')|^2 \widetilde{F}_a^2\left(2\frac{\omega'}{c}\sin\left(\frac{\theta}{2}\right)\right), \qquad (9)$$

here θ is the scattering angle. The expression for the angular distribution of the scattering probability follows directly from eq. (9):

$$\frac{dW}{d\Omega'} = \frac{1+\cos^2\theta}{8\pi^2 c^3} \int_0^\infty Z_{pol}^2(\omega') E_0^2 |f(\omega')|^2 \widetilde{F}_a^2\left(2\frac{\omega'}{c}\sin\left(\frac{\theta}{2}\right)\right)\frac{d\omega'}{\omega'}. \qquad (10)$$

The total scattering probability is given by

$$W(\tau,\omega) = \frac{2}{3\pi c^3} \int_0^\infty Z_{pol}^2(\omega') E_0^2 |f(\omega')|^2 D_a(\omega')\frac{d\omega'}{\omega'}, \qquad (11)$$

where

$$D_a(\omega') = \frac{3}{8}\int_0^\pi \left(1+\cos^2\theta\right)\widetilde{F}_a^2\left(2\frac{\omega'}{c}\sin\left(\frac{\theta}{2}\right)\right)\sin\theta\, d\theta \qquad (12)$$

describes the decrease of the elastic scattering probability due to the non-dipole interaction between the radiation and the atom. In the low-frequency limit (dipole approximation) $D_a(\omega') \rightarrow 1$. The non-dipole function of eq. (12) is presented in Fig. 2 for Mg, Al and Fe atoms in XUV and soft X-ray spectral ranges.
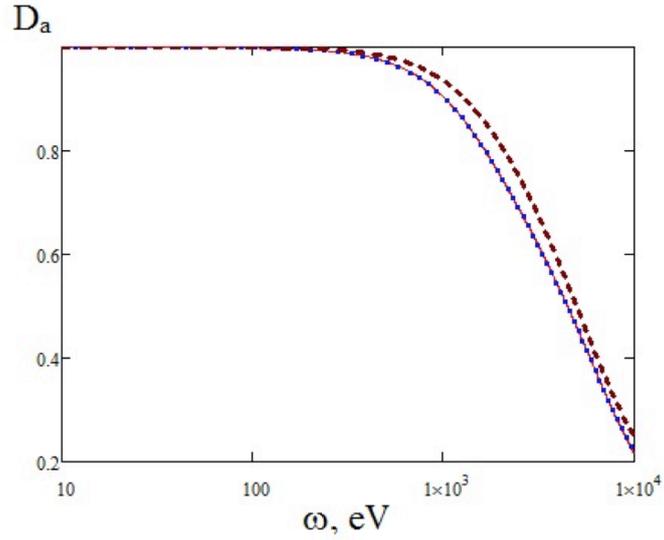
Figure 2: *Non-dipole interaction function D_a for Mg (solid red line), Al (dotted blue line) and Fe (dashed brown line) atoms.*

In XUV range ($\omega < 100$ eV) the influence of non-dipole interaction is negligible and the atomic form factor $\widetilde{F}_a(q)$ and the non-dipole scattering term $D_a(\omega')$ can be set equal to unity in eqs. (9) - (11).

We now consider the properties of the polarization charge and investigate the USP scattering on Mg, Al and Fe atoms in the photon energy range above 30 eV. The spectra of polarization charge of these atoms calculated according relation (4) employing the data from [21] are presented in Fig. 3.

(a)
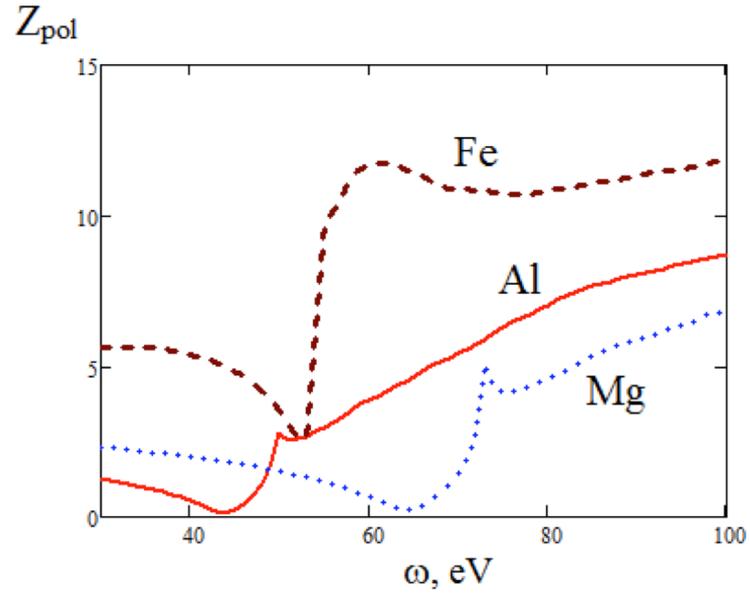
(b)
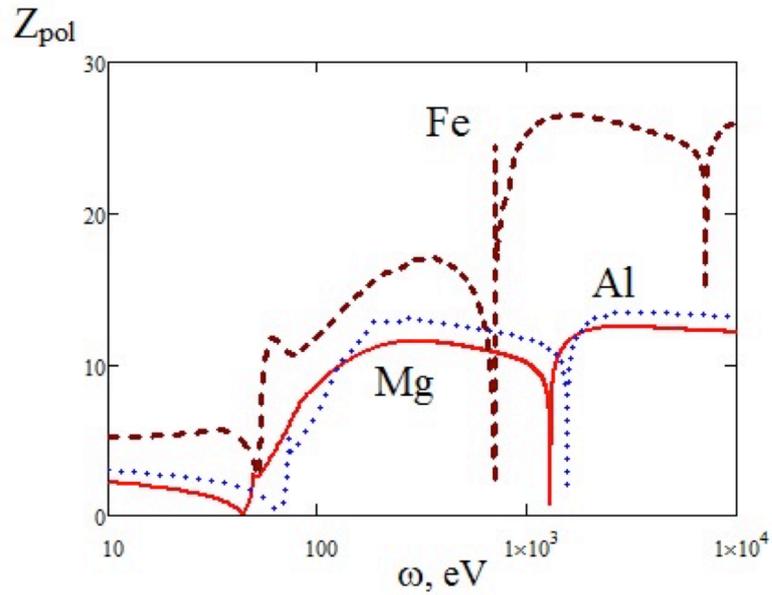
Figure 3: *Polarization charges of Mg (solid red line), Al (dotted blue line) and Fe (dashed brown line) atoms as a function of frequency in XUV (a) and soft X-ray (b) spectral ranges.*

It is apparent from Figs. 3a,b that polarization charges have asymmetric minima as a function of frequency. The positions of these minima are correlated with the ionization potential of atomic shells [22] and are located in the vicinity of the *2p* shell for Mg and Al and the *3p* shell for Fe.

III. Results: non-linear scattering probability and resonance like features

For the following calculations of attosecond pulse scattering on matter we use the Gaussian pulse (GP) shape in the following form:

$$f(\omega') = i\tau\sqrt{\frac{\pi}{2}}\left[\exp\left(-(\omega-\omega')^2\tau^2/2\right) - \exp\left(-(\omega+\omega')^2\tau^2/2\right)\right], \qquad (13)$$

Here τ and ω are duration and carrier frequency of the pulse. This shape function is the Fourier transform of the electric field of the incident pulse depending on the retarded time:

$$E(t_r) = \exp\left(-t_r^2/2\tau^2\right)\sin(\omega t_r) \qquad (14)$$

Below we present results of numerical calculations for the scattering probability for USP on Mg, Al and Fe atoms for $E_0=1$ atomic unit in order to provide numerical data below which the respective asymptotic limits are fulfilled (this relates likewise to the possibility for experimental studies and diagnostics). Let us consider firstly the spectrum of the scattering probability. Integration of eq. (9) over the scattering angles provides:

$$\frac{dW}{d\omega'} = \frac{2}{3\pi c^3\omega'}Z_{pol}^2(\omega')E_0^2\left|f(\omega')\right|^2 D_a(\omega'), \qquad (15)$$

here ω' is the frequency of scattering radiation. The spectra of eq. (15) of ultra-short GP scattered on Al atoms are presented in Fig. 4a-c (solid lines) for different pulse durations: (a) $\tau=20$ as, (b) $\tau=80$ as and (c) $\tau=500$ as at a carrier frequency equal to electron binding energy of the 2p subshell ($\omega_{Al,2p}=72.7$ eV). For comparison the normalized spectrum of the squared polarization charge divided by frequency and the normalized spectrum of incident pulses are also shown.

Figure 4 shows that the spectrum of the scattered pulse is greatly distorted in comparison with the spectrum of the incident pulse in frequency range near the sharp dip of atomic polarization charge and for short pulse duration. Calculations shows that similar distortion of the scattered spectra occurs for other atoms, e.g. Fe the spectrum considerably changes for somewhat longer pulses in comparison with Mg and Al due to narrower minimum in the spectrum of the polarization charge. It follows from Fig. 4 that for sufficiently short pulses the scattered spectrum reproduces the squared polarization charge spectrum (divided by frequency) for a carrier frequency that is equal to the electron binding energy and in spectral range where the polarization charge spectrum has a sharp dip.

(a)
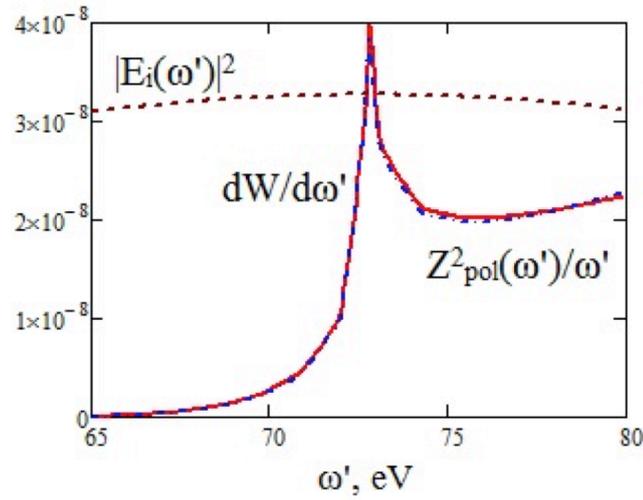
(b)
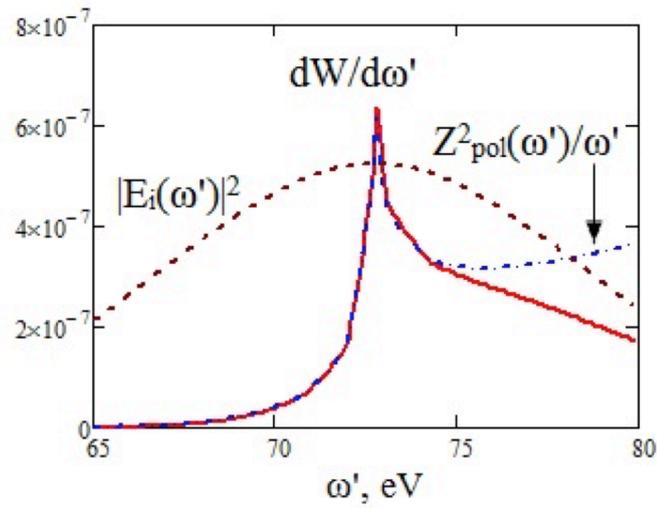
(c)
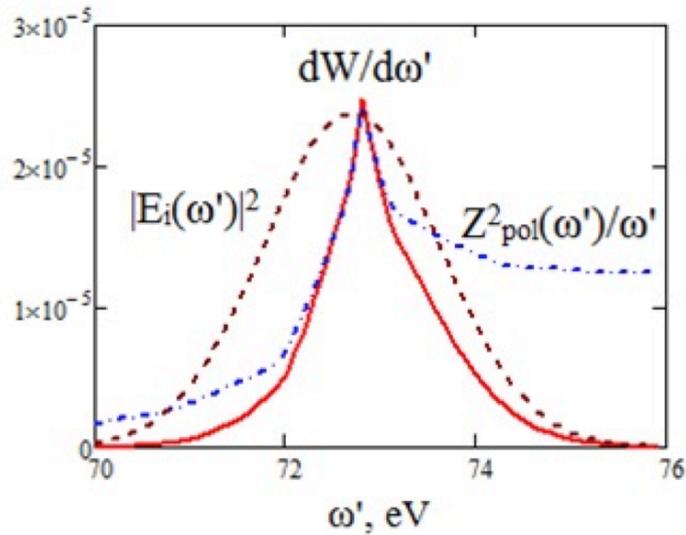
Figure 4: *Solid red lines - spectra of GP scattering on Al atom for pulse durations of 20 as (a), 80 as (b), 500 as (c) and carrier frequency 72.7 eV; dashed brown lines – spectra of incident pulses (multiplied by numerical factor $3 \cdot 10^{-8}$ (a,b) and $3.5 \cdot 10^{-8}$ (c)), dotted-dashed blue lines – squared polarization charge of Al atom divided by frequency and multiplied by numerical factors $8.75 \cdot 10^{-8}$ (a), $1.4 \cdot 10^{-6}$ (b), $5.5 \cdot 10^{-5}$ (c)*
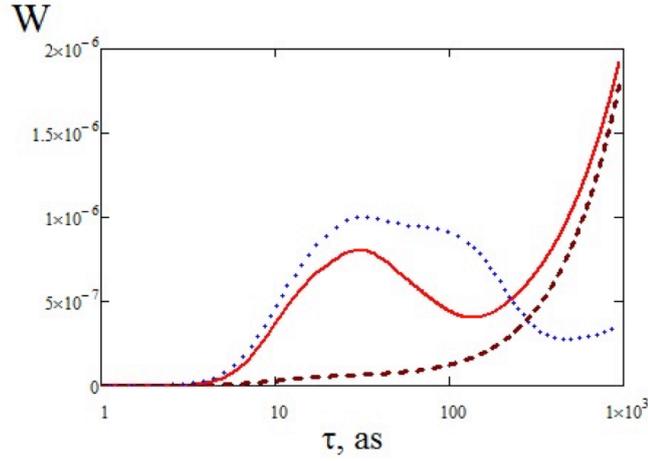
Figure 5: *Total scattering probability of GP on Mg atom as a function of pulse duration for different carrier frequencies near the first minimum of $Z_{pol}(\omega)$: solid red line – ω=40 eV, dotted blue line – ω=45 eV, dashed brown line – ω=50 eV (for the latter case the scattering probability is multiplied by a factor of 0.05).*
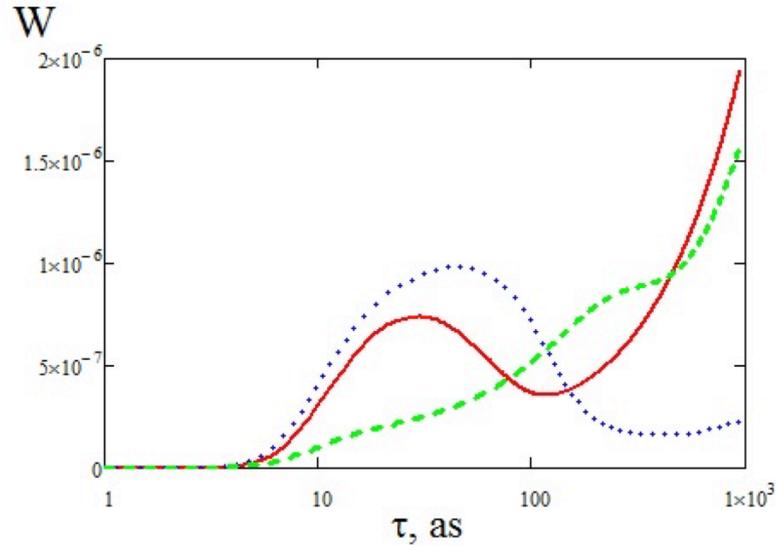

Fig.6. Total scattering probability of GP on Al atom as a function of pulse duration for different carrier frequencies near the first minimum of $Z_{pol}(\omega)$: solid red line – ω=60 eV, dotted blue line – ω=65 eV, dashed green line – ω=70 eV (for the latter case the scattering probability is multiplied by a factor of 0.2).

Figures 5-7 demonstrate that for specific carrier frequencies the total probabilities of GP scattering are non-linear functions of pulse duration. These respective frequency ranges correspond to minima in the spectra of polarization charges of the considered atom. As one can see from Fig. 3a above mentioned minima correspond to the following photon energies (in XUV

range): 45 eV for Mg, 65 eV for Al and 52.5 eV for Fe and in soft X-ray range (Fig 3b): 1302 eV for Mg, 1559 eV for Al and 700 eV for Fe. It should be noted that the frequencies of these minima are somewhat lower than the values of the respective ionization potentials.
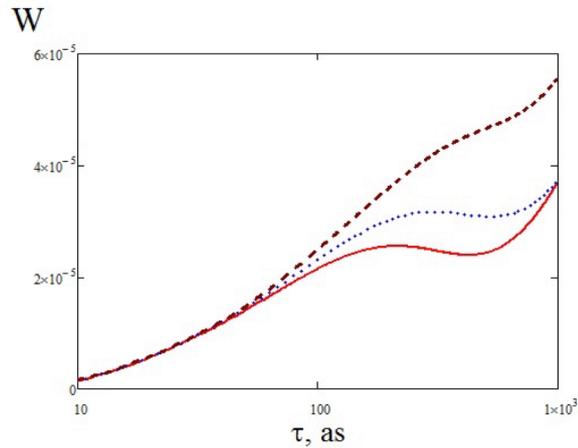

Figure 7: *Total scattering probability of GP on Fe atom as a function of pulse duration for different carrier frequencies near the first minimum of $Z_{pol}(\omega)$: solid red line – ω=52 eV, dotted blue line – ω=52.5 eV. dashed brown line – ω=53 eV.*

Figs. 5-7 demonstrate that extrema of W(τ) shift somewhat to longer pulse durations for carrier frequencies that are equal to the spectral position of the polarization charge minima.
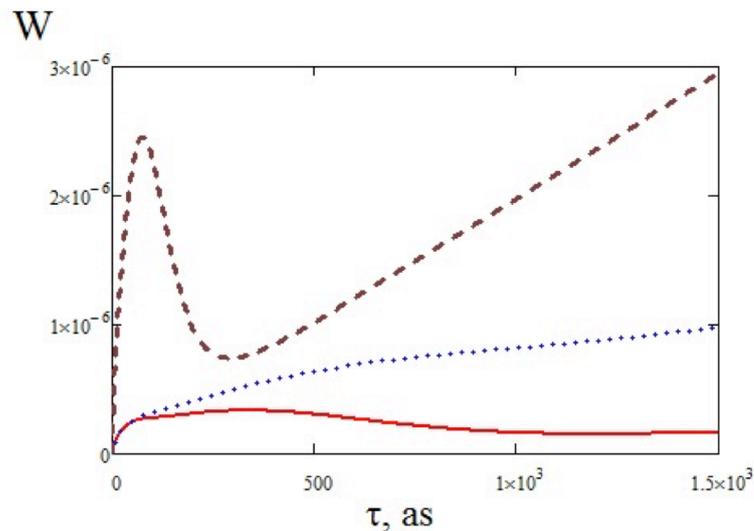

Figure 8: *Total scattering probability of GP on Mg (solid red line), Al (dotted blue line) and Fe (dashed brown line) atoms as a function of pulse duration for carrier frequencies near the second sharp minima of $Z_{pol}(\omega)$ (see Fig. 3b) in soft X-ray range: solid red line – ω=1302 eV, dotted blue line - ω=1559 eV, dashed brown line - ω=700 eV.*

Figure 8 demonstrates the dependence of W(τ) for Mg, Al and Fe for carrier frequencies that correspond to the second minima in spectra of the polarization charges (Fig. 3b). In this case extrema for Mg considerably shift to longer pulse durations but are not so pronounced as in the XUV range (see Fig. 5). For Al atoms extrema are absent and for Fe atoms the positions of extrema shift in short pulse duration range. Table 1 summarizes the positions of extrema for the W(τ)-function.

Table 1: Position of Extrema of the scattering probability W(τ)

atom	ω_1, eV	τ_{1max}, as	τ_{1min}, as	ω_2, eV	τ_{2max}, as	τ_{2min}, as
Mg	45	30-110	540	1302	350	1200
Al	65	50	440	1559	-	-
Fe	52.5	300	600	700	80	285

As follows from eq. (10) the shape of the angular distribution of the scattering probability is practically independent upon the USP duration for all frequencies and has the same form as for long pulses.

IV. Conclusion

The peculiarities of attosecond pulse scattering on atoms are investigated in XUV and soft X-ray ranges when the atomic state does not change during the process (elastic scattering). Unlike the long pulse limit where the scattering probability per unit time is relevant, we study the dependence of the scattering probability for the total duration of ultra-short pulse action on atoms that results in highly non-linear and even resonance like features. Numerical calculations are carried out for Mg, Al and Fe atoms for pulse duration of 10-100 attoseconds that employ an efficient non-dipole scattering tensor approximation that expresses the scattering probability in terms of frequency dependent polarization charge of an atom and the atomic form-factor. It is shown that the presence of sharp minima in spectrum of the atomic polarization charge leads to the non-linear dependence of the total scattering probability upon the pulse duration W(τ) in contrast with long pulse limit when the scattering probability is linear function of pulse duration τ. It is shown that sufficiently narrow minima in the spectrum of the polarization charge result in extrema of the scattering probability W(τ) in dependence of the attosecond pulse duration. The positions of these extrema shift to larger values of USP duration with decreasing width of minima in the polarization charge spectrum.

The calculations demonstrate that the spectrum of USP scattering significantly changes for carrier frequencies near the minima of atomic polarization charge. It is shown that

measurements for pulses that are sufficiently close to their respective short pulse limit can be used for the determination of squared polarization charge spectrum, associated atomic characteristics such as scattering cross sections and dynamical polarizability of an atom in the most interesting spectral range where resonance like features dominate.

V. Acknowledgement

VI. References

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