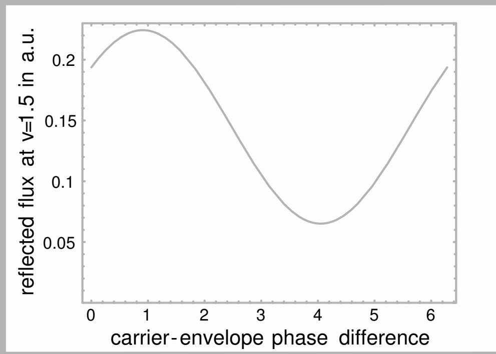
Laser Phys. Lett. **1**, No. 1, 42–45 (2004) / **DOI** 10.1002/lapl.200310010

Abstract: On the basis of classical electrodynamics the reflection and transmission of a few-cycle femtosecond Ti:Sa laser pulse impinging on a thin metal layer have been analysed. The thickness of the layer was assumed to be much smaller than the skin depth of the radiation field, and the metallic electrons were represented by a surface current density. The interaction of the electrons with a periodic lattice potential has also been taken into account. The presence of this nonlinear potential leads to the appearance of higher harmonics in the scattered spectra. A formal exact solution has been given for the system of the coupled Maxwell-Lorentz equations describing the dynamics of the surface current and the radiation field. Besides, an analytic solution was found in the strong field approximation for the Fourier components of the reflected and transmitted radiation. In our analysis particular attention has been paid to the role of the carrier-envelope phase difference of the incoming few-cycle laser pulse.



Shows the dependence on the carrier-envelope phase difference of the reflected flux of a two-cycle Ti:Sa laser pulse at the first relative minimum of the spectrum (v=1.5)

Scattering of a few-cycle laser pulse on a thin metal layer : the effect of the carrier-envelope phase difference

S. Varró *

Research Institute for Solid State Physics and Optics, H-1525 Budapest, POBox 49, Hungary

Received: 14 October 2003, Accepted: 24 October 2003
Published online: 17 December 2004

Key words: few-cycle laser pulses; high harmonic generation; carrier-envelope phase difference

PACS: 41.20 Jb, 42.25 Bs, 52.38-r

1. Introduction

The effect of the absolute phase on the nonlinear response of atoms and solids interacting with a very short, few-cycle strong laser pulse has recently drawn considerable attention and has initiated a wide-spreading theoretical and experimental research [1–7]. The theoretical works have appeared by now on the ionization use the quantum description of the interacting electrons either bound to an atom [4–6] or in a metal layer [7]. In the present paper we briefly describe our theoretical analysis on the reflection and transmission of a few-cycle laser pulse on a thin metal layer represented by a surface current.

Our analysis is completely based on classical electrodynamics and mechanics, in the frame of which we solve the system of coupled Maxwell-Lorentz equations of the incoming and scattered radiation and the classical surface current. The idea to study such a system appeared to us by reading a paper by Sommerfeld [8] published in Analen der Physik in 1915 in which he analysed the temporal distortion of x-ray pulses impinging perpendicularly on a surface current being in vacuum. We have generalized this model in the following sense. On one hand, we allow oblique incidence of the incoming radiation field, and on the other hand, we assume that the surface current (which represents a thin metal layer) is embedded to two semi-infinite dielectrics with two different index of refractions. Moreover, in the equation of motion for the electrons we introduce the interaction with a periodic lattice potential represented by a cosine function. The presence of this nonlinear potential is responsible for the appearance of high-harmonics both in the reflected and transmitted radiation.

In Section 2 we present the basic equations describing our model. In Section 3 we give a short discussion of a

* Corresponding author: e-mail: varro@sunserv.kfki.hu

representative scattered spectrum and show how the intensity depends on the carrier-envelope phase difference of the incoming few-cycle laser pulse.

2. The basic equations of the model

We take the coordinate system such that the first dielectric with index of refraction n_1 fills the region $z > l_2/2$, this is called region 1. In region 2 we place the thin metal layer of thickness l_2 perpendicular to the z-axis and defined by the relation $-l_2/2 < z < +l_2/2$. Region 3, $z < -l_2/2$, is assumed to be filled by the second dielectric having the index of refraction n_3. The thickness l_2 is assumed to be much smaller then the skin depth of the incoming radiation. The target defined this way can be imagined as a thin metal layer evaporated, for instance, on a glass substrate. In case of perpendicular incidence the light would come from the positive z-direction, and it would be transmitted in the negative z-direction into region 3. The plane of incidence is defined as the y-z plane and the initial **k**-vector is assumed to make an angle θ_1 with the z-axis. In case of an s-polarized incoming TE wave the components of the electric field and the magnetic induction read $(E_x, 0, 0)$ and $(0, B_y, B_z)$, respectively. They satisfy the Maxwell equations

$$\partial_y B_z - \partial_z B_y = \partial_0 \varepsilon E_x, \tag{1}$$

$$\partial_z E_x = -\partial_0 B_y, -\partial_y E_x = -\partial_0 B_z,$$

where $\varepsilon = n^2$ is the dielectric constant and n is the index of refraction. If we make the replacements $\varepsilon E_x \to -B_x, B_z \to E_z$ and $B_y \to E_y$ then we have the field components of a p-polarized TM wave $(0, E_y, E_z)$and $(B_x, 0, 0)$, and we receive the following equations

$$\partial_z B_x = \partial_0 \varepsilon E_y, -\partial_y B_x = \tag{2}$$

$$= \partial_0 \varepsilon E_z, \partial_y E_z - \partial_z E_y = -\partial_0 B_x.$$

In the followings we will consider only the latter case, namely the scattering of a p-polarized TM radiation field. From Eq. (2) we deduce that B_x satisfies the wave equation, and in region 1 we take it as a superposition of the incoming plane wave pulse $F$and an unknown reflected plane wave f_1

$$B_{x1} = F - f_1 = F[t - n_1(y \sin\theta_1 - z\cos\theta_1)/c] - \tag{3}$$

$$-f_1[t - n_1(y\sin\theta_1 + z\cos\theta_1)/c].$$

From Eq. (2) we can express the components E_y and E_z of the electric field strength by taking into account Eq. (3)

$$E_{y1} = (\cos\theta_1/n_1)(F + f_1), E_{z1} = \tag{4}$$

$$= (\sin\theta_1/n_1)(F - f_1).$$

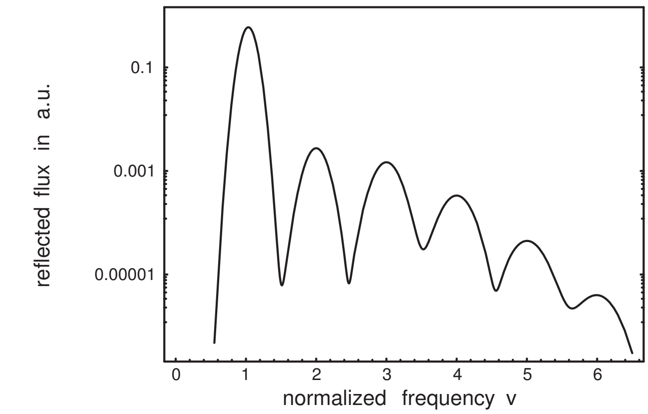


Figure 1 Shows the spectrum of the reflected two-cycle Ti:Sa laser pulse of intensity 10^{13} W/cm^2 impinging at 45 degree of incidence on an aluminum layer of thickness $\lambda_0/100$

In region 3 the general form of the magnetic induction B_{x3} is the by now unknown refracted wave g_3

$$B_{x3} = g_3 = g_3[t - n_3(y\sin\theta_3 - z\cos\theta_3)/c]. \tag{5}$$

The corresponding components of the electric field strength are expressed from the above equation with the help of the first two equation of (2)

$$E_{y3} = (\cos\theta_3/n_3)g_3, E_{z3} = (\sin\theta_3/n_3)g_3. \tag{6}$$

In region 2 the relevant Maxwell equations with the current density **j** read

$$\partial_z B_x = (4\pi/c)j_{y2} + \partial_0 \varepsilon E_y, \tag{7}$$

$$\partial_y E_z - \partial_z E_y = -\partial_0 B_x.$$

By integrating the two equations in (7) with respect to z from $-l_2/2$ to $+l_2/2$ and taking the limit $l_2 \to 0$, we obtain the boundary conditions for the field components

$$[B_{x1} - B_{x3}]_{z=0} = (4\pi/c)K_{y2}, [E_{y1} - E_{y3}]_{z=0} = 0, \tag{8}$$

where K_{y2} is the y-component of the surface current in region 2. This surface current can be expressed in terms of the local velocity of the electrons in the metal layer

$$K_{y2} = e(d\delta_{y2}/dt)l_2 n_{e2}, \tag{9}$$

$$(4\pi/2c)K_{y2} = (m/e)\mathit{\Gamma}_2(d\delta_{y2}/dt),$$

$$\mathit{\Gamma}_2 \equiv 2\pi(e^2/mc)l_2 n_{e2}, \tag{10}$$

$$\mathit{\Gamma}_2 = (\omega_{p2}/\omega_0)^2(\pi l_2/\lambda_0)\omega_0,$$

where for later convenience we have introduced ω_0, $\lambda_0 = 2\pi c/\omega_0$, the carrier frequency and wavelength of the incoming light pulse, and n_{e2}, $\omega_{p2} = \sqrt{4\pi n_{e2}e^2/m}$, the density of electrons and the corresponding plasma frequency in the metal layer, respectively. In Eq. (9) δ_{y2} denotes the local displacement of the electrons in the metal layer for which we later write down the Newton equation in the presence of the complete electric field. We remark that in reality the thickness l_2 is, of course, not infinitesimally small, rather, it has a finite value which is anyway assumed to be smaller then the skin depth $\delta_{skin} \approx c/\omega_{p2}$. For instance, for an aluminum layer of thickness $l_2 \approx \lambda_0/100$we have $\Gamma_2 \approx 3\omega_0$.

From Eq. (8) with the help of Eq. (9) we can express f_1 and g_3 in terms of $\delta'_{y2}(t')$

$$f_1(t') = \qquad (11)$$
$$= (1/(c_1+c_3))[(c_3-c_1)F(t') - 2c_3(m/e)\Gamma_2\delta'_{y2}(t')],$$

$$g_3(t') = (2c_1/(c_1+c_3))[F(t') - (m/e)\Gamma_2\delta'_{y2}(t')], \quad (12)$$

where the prime on δ_{y2} denotes the derivative with respect to the retarded time $t' = t - yn_1 \sin\theta_1/c$ which is equal to $t - yn_3 \sin\theta_3/c$, securing Snell's law of refraction $n_1 \sin\theta_1 = n_3 \sin\theta_3$ to hold. Moreover, in Eqs. (11) and (12) we have introduced the notations

$$c_1 = \cos\theta_1/n_1,\, c_3 = \cos\theta_3/n_3. \quad (13)$$

The equation of motion of δ'_{y2} can be easily derived from Eqs. (4) and (11) yielding

$$\delta''_{y2}(t') = (2c_1c_3/(c_1+c_3))[(e/m)F(t') - \qquad (14)$$
$$-\Gamma_2\delta'_{y2}(t')] + (e/m)F_U[\delta_{y2}(t')].$$

In the above equation we have introduced the force term $F_U = -\partial U/\partial\delta_{y2}$ which represents the effect of the ionic cores in the metal layer, where $U = U_0\cos(\kappa\delta_{y2}+\chi)$ approximates the lattice potential. Typically $U_0 \sim 0.1 - 1$Volts and $\kappa \sim 2\pi/$ Angström. For definiteness, we impose the initial conditions on the electron displacement $\delta_{y2}(-\infty) = 0$ and $\delta'_{y2}(-\infty) = 0$. Owing to Eqs. (11) and (12) the solution of Eq. (14) gives at the same time the complete solution of the scattering problem. Without the term $\propto F_U$, Eq. (14) can formally be solved exactly for an arbitrary incoming field $F(t)$, by using Fourier transformation technique.

3. Exact formal and approximate analytic solution of the scattering problem

By calculating the Fourier transforms of Eqs. (11), (12) and (14) we can give an exact formal solution of the scattering problem, namely

$$f_1(\omega) = -\frac{[F(\omega)+F_U(\omega)/b]}{\gamma - i(v/b)}\left[\gamma + \frac{c_3-c_1}{c_3+c_1}i(v/b)\right], \quad (15)$$

$$g_3(\omega) = -\frac{2c_1}{c_1+c_3}\frac{i(v/b)[F(\omega)+F_U(\omega)/b]}{\gamma - i(v/b)}, \quad (16)$$

where $\gamma \equiv \Gamma_2/\omega_0$, $v \equiv \omega/\omega_0$ and $b \equiv 2c_1c_3/(c_1+c_3)$. It can be proved that the Fourier components of the reflected and the transmitted fluxes satisfy the following sum rule

$$c_1|f_1(\omega)|^2 + c_3|g_3(\omega)|^2 = c_1|F(\omega)+F_U(\omega)/b|^2. \quad (17)$$

By now we have not specified the explicit form of the incoming field. Now let us assume that it is a gaussian quasi-monochromatic field with a carrier frequency ω_0 having the carrier-envelope phase difference φ. We derive this field from the Hertz potential $Z(t)$ in the usual way

$$F(t) = -(1/c^2)\partial^2 Z(t)/\partial t^2,\, Z(t) = \qquad (18)$$
$$= (c^2/\omega_0^2)F_0\exp(-t^2/2\tau^2)\cos(\omega_0 t+\varphi),$$

where $\tau = \tau_L/2\sqrt{\log 2}$ with τ_L being the full temporal width at half maximum of the pulse. In zeroth approximation we can neglect the effect of the lattice field F_U and the Newton equation (14) with the only force term (18) can be analytically solved giving the approximate trajectory $\delta^{(0)}_{y2}(t')$

$$\delta^{(0)}_{y2}(t) \approx \qquad (19)$$
$$\approx -b\frac{e}{m\omega_0^2}\frac{F_0}{\sqrt{1+b^2\gamma^2}}e^{-t^2/2\tau^2}\cos(\omega_0 t+\varphi+\eta),$$
$$\sin\eta = \frac{b\gamma}{\sqrt{1+b^2\gamma^2}},$$

where γ and b were already defined after Eq. (16).

By inserting this trajectory into the argument of $F_U[\delta^{(0)}_{y2}(t')]$, the Fourier transform $F_U(\omega)$ can be approximately calculated yielding an infinite sum of terms peaked at the higher harmonic frequencies $\omega = n\omega_0$ with $n = 1, 2, 3, ...$

$$F_U(\omega) \approx -i\cos\chi F_{U0}\tau\times \qquad (20)$$
$$\times\sum_{n=1,3,...}\frac{J_n(a)}{\sqrt{2\pi n}}\{\exp[-\varsigma - \tau^2\omega_0^2((\omega/\omega_0)+n)^2/2n] -$$
$$-\exp[\varsigma - \tau^2\omega_0^2((\omega/\omega_0)-n)^2/2n]\} + \sin\chi F_{U0}\tau\times$$
$$\times\sum_{n=2,4,...}\frac{J_n(a)}{\sqrt{2\pi n}}\{\exp[-\varsigma - \tau^2\omega_0^2((\omega/\omega_0)+n)^2/2n] +$$
$$+\exp[\varsigma - \tau^2\omega_0^2((\omega/\omega_0)-n)^2/2n]\},$$
$$\varsigma = in(\varphi+\eta+\pi/2),$$

where $J_n(a)$ denotes an ordinary Bessel function of first kind of order n and

$$a \equiv -b\frac{\kappa e}{m\omega_0^2}\frac{F_0}{\sqrt{1+b^2\gamma^2}}. \quad (21)$$

The Fourier transform of the incoming pulse given by Eq. (18) reads

$$F(\omega) = (\omega^2/2\omega_0^2)(F_0\tau/\sqrt{2\pi})\times \qquad (22)$$
$$\times\{\exp(-i\varphi)\exp[-\tau^2\omega_0^2((\omega/\omega_0)+1)^2/2] +$$
$$+\exp(+i\varphi)\exp[-\tau^2\omega_0^2((\omega/\omega_0)-1)^2/2]\}.$$

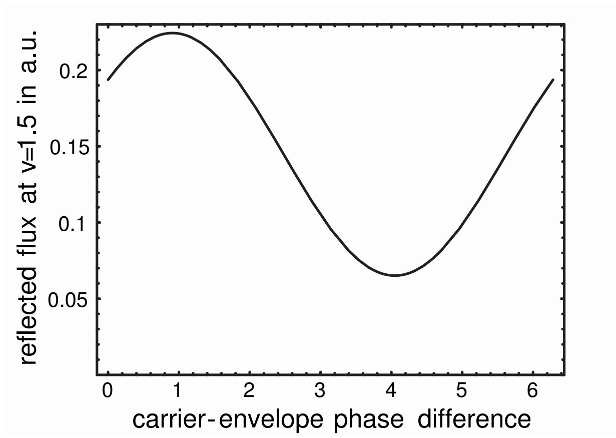


Figure 2 Shows the dependence on the carrier-envelope phase difference of the reflected flux of a two-cycle Ti:Sa laser pulse at the first relative minimum of the spectrum (v=1.5)

We give a numerical example on one hand for the dependence of the reflected flux (see Figure 1) on the normalized frequency $v = \omega/\omega_0$, and, on the other hand, on the φ-dependence of the flux at $v = 1.5$.

As can be seen on Fig. 2 the modulation of the reflected flux is quite deep halfway at the fundamental and the second harmonic where $v = \omega/\omega_0 = 1.5$. The modulation function $M(\omega) = (I_{\max} - I_{\min})/(I_{\max} + I_{\min})$ is about 64% in this case. We have checked that as we increase the pulse duration M becomes practically zero if the pulse contains more than five cycles. Unfortunately, even for few-cycle pulses, we have not observed any dependence on the carrier-envelope phase difference at the maxima of the spectrum. Considerable modulation always appears at the local minima of the spectrum, i.e. at $v = 1.5, 2.5, ...$ and rapidly drops to zero in the vicinity of this points. The physical reason for this phenomenon is that the modulation stems from the interference terms appearing in $|F(\omega) + F_U(\omega)/b|^2$.

Acknowledgements This work has been supported by the Hungarian National Scientific Research Foundation (OTKA, grant no. T032375), by European Center-of-Excellence Program (KFKI Condensed Matter Research Center, contract no. ICAI-CT-2000-70029) and by the Scientific-Technical Collaboration Agreement between Hungary and Austria (2002/2003, project no. A-1/01).

References

[1] T. Brabec and F. Krausz, Rev. Mod. Phys. **72**, 545–591 (2000).

[2] G.G. Paulus, F. Grasbon, H. Walther, P. Villoresi, N. Nisoli, S. Stagira, E. Priori, and S. De Silvestri, Nature **414**, 182–185 (2001).

[3] M. Hentschel, R. Kienberger, Ch. Spielmann, G.A. Reider, N. Milosevic, T. Brabec, P. Corkum, and F. Krausz, Nature **414**, 509–513 (2001).

[4] S. Chelkowski and A.D. Bandrauk, Phys. Rev. A **65**, 061802(R)(1–4) (2002).

[5] D.B. Milosevic, G.G. Paulus, and W. Becker, Phys. Rev. Lett. **89**, 15300(1–4) (2002).

[6] D.B. Milosevic, G.G. Paulus, and W. Becker, Optics Express **11**, 1418–1429 (2003).

[7] Ch. Lemell, X.-M. Tong, F. Krausz, and J. Burgdörfer, Phys. Rev. Lett. **90**, 076403(1–4) (2003).

[8] A. Sommerfeld, Ann. d. Physik **46**, 721–747 (1915).