Time-resolved interference microscopy for studying nonideal plasma formed by high-power femtosecond laser pulses

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Abstract. Measurement results of characteristic size of plasma density inhomogeneity on the surface of bulk iron target formed under action of femtosecond laser pulses with intensity of $10^{16}$ W/cm$^2$ are presented. Investigations are performed using time-resolved interference microscopy technique. Femtosecond laser pulses with high time contrast of $10^7$ generated by chromium:forsterite laser system are applied. Efficacy of the chosen technique is demonstrated; the size of plasma inhomogeneity is shown to be less than 30 nm.

1. Introduction
Rapid development of physics and technologies has made possible laser systems with unique characteristics to emerge-tremendous intensity ($I > 10^{21}$ W/cm$^2$), high contrast (up to $10^{15}$) [1], and short duration (several tens of femtoseconds) [2-6]. Investigation of interaction of laser radiation with such intensities with matter is not only of fundamental, but also of practical importance, ranging from generation of X-ray and gamma radiation to ion acceleration in cancer therapy [7-9], etc.

To generate efficiently characteristic X-ray radiation, electrons with appropriate energies are required. To accelerate electrons, not only structured targets (for example, nanowires [10] or grating targets for generation of propagating surface plasmons on their surface [11]) are used, but also parameters of laser pulse (chirp [12] and polarization [13]) are manipulated. Methods for modeling laser-plasma interaction processes are also constantly being improved; about a dozen of theoretical papers on this topic have been published in 2018 only (for example, see [14, 15]).

At the same time, correct simulation of the considered processes requires experimental data to validate itself, since plasma properties on the target surface depend not only on characteristics of the latter (structure, composition, thickness and density), but also on such parameters of laser pulse as duration, energy, power density, and polarization. As intensity of laser pulse increases, its time profile comes first, since the presence of a pre-pulse can lead to formation of a pre-plasma on the target surface, which affects the efficiency of fast (or hot) electron generation [16, 17]. In these papers, the effect of pre-plasma on both the generation processes of fast electrons (spectrum, spatial distribution, and angular divergence) and their temperature was studied. At intensities of a laser pulse, for which the condition $I\lambda^2 \leq 10^{18}$ W/cm$^2$ $\mu$m$^2$ is satisfied, generation of hot electrons can be caused mainly by two mechanisms: the vacuum heating and the resonant absorption. Numerical simulation by means of particle-in-cell (PIC) method [18] has shown that vacuum heating can be more efficient than resonant absorption in many cases.
To assess the effect of a particular mechanism, X-ray characteristic radiation generated in plasma due to impact of a $p$-polarized femtosecond pulse is used as a diagnostic tool. Earlier in [19], to determine the prevailing mechanism of hot electrons generation, experimental dependences were obtained of intensity of $K_a$ radiation on the angle of incidence of laser pulse on the target surface. However, the obtained results have demonstrated that generation of hot electrons in the whole range of angles can be due to both of the above mechanisms; the decisive role is played by the characteristic size of the plasma density inhomogeneity in the vicinity of the critical density.

In this paper a method of time-resolved interferometric microscopy is proposed for the first time to study the size of pre-plasma on the target surface when exposed to a femtosecond laser pulse.

2. Experimental setup

Similar to previous study [19], an infrared terawatt femtosecond laser system based on the chromium:forsterite active element is used to generate X-ray characteristic radiation. It provides laser pulses with an energy of up to 90 mJ and a repetition rate of 10 Hz, $\sim 80 \div 20$ fs in duration at a wavelength of 1240 nm and a spectral width of 26 nm (FWHM) [4].

Time-resolved interference microscopy technique [20] is used to study the dynamics of preplasma formation and expansion as a result of the impact of femtosecond pulse with an intensity of $10^{16}$ W/cm$^2$. The applied approach allows to obtain information on the changes of amplitude $r_{ind}$ and phase $\Psi_{ind}$ of the complex reflection coefficient. Previously, this technique has been used to study such processes as the formation of electron-hole plasma in semiconductors [21], phase transitions in solids [22], laser ablation of the surface under the action of ultrashort laser pulses [23], and determining the optical and transport properties of plasma at an laser intensity of $10^{15}$ W/cm$^2$ [24], as well as to determine the strength of metals in the liquid phase [25, 26]. It is based on the pump-probe scheme, in which laser radiation is divided into two beams – the pump (more powerful) and the probe ones. To change the energy of laser pulses, a polarization attenuator (a half-wave plate and a prism polarizer) is installed in each arm, and the energy is controlled using a calibrated germanium photodiode. The probe pulse ($\lambda = 620$ nm, frequency is doubled in DKDP crystal) is used to illuminate the sample area under study. Standa 8MT160-300 delay-line used in the scheme allows changing the time delay $t_{\text{delay}}$ between the pump and the probe pulses with 8 fs steps. The uniformity of illumination of the sample area is reached using a spatial filter assembled in the probe beam; it is a Kepler 1:1 telescope with a diaphragm 30 µm in diameter installed in the common focal plane of the lenses.

Michelson interferometer is assembled in a vacuum chamber. The chamber has four flanges, including optical windows, electrical connectors, and inlet valves. Laser radiation comes through the input window with antireflective coating for both the fundamental and the second harmonic frequencies of laser radiation. The evacuation of the chamber is performed up to a pressure of $\sim 10^{-3}$ Torr.

The image transfer of the target surface to the CCD plane (1024 × 1024 pixels) with magnification M $\sim 30$ is performed using a micro-lens 9×, NA = 0.2. The second arm of the interferometer is formed by a micro-lens with similar parameters and a reference mirror. To align the intensity in the interferometer arms, a set of neutral optical filters installed between the reference mirror and the micro-lens is used. The probe beam reflected from the target surface (called "object" beam) interferes with the "reference" beam in the CCD plane of the camera placed outside the vacuum chamber. Application of CCD camera (SensiCam QE, PCO) with 12-bit depth allows interferograms to be recorded with a smaller quantization step of intensity than common 8-bit cameras that increases the sensitivity of the experimental setup. The intrinsic plasma emission is cut off by a set of optical filters consisting of a narrow-band interference filter with transmission at a wavelength of the probe pulse $\lambda = 620 \pm 10$ nm, as well as SZS-23, KS-11 and OC-4 ones. A couple of interferograms of the target surface are recorded in the experiment – prior to laser impact and at the moment of laser irradiation (at a certain $t_{\text{delay}}$ value). Processing of these interferograms [20] results in spatial distribution of the amplitude and phase changes of the complex reflection coefficient. Further, only the phase change
\[ \psi_{\text{ind}}(x, y) \] of the reflection coefficient will be of interest. Its accuracy in interference microscopy setup is up to \( \Delta \psi_{\text{ind}} \approx 0.01 \text{ rad} \).

Similar to [23], the target is a bulk cylindrical sample of iron (30 mm in diameter and 35 mm in height) with a side surface roughness not worse than \( R_z = 10 \). The target is set on a 3D node consisting of three motorized linear translators (Standa 8MT-173-20) and a rotation one (Standa 8MR-174-11). The target is oriented so that the probe laser pulse falls normally the cylindrical surface and the pump one has 45º angle of incidence. The pulp pulse is focused by an off-axis parabolic mirror with a focal length of 2\( " \) (Thorlabs MPD229-M01) 2\( " \) in diameter.

![Figure 1](image1.png)

**Figure 1.** Time-resolved interference microscopy scheme. 1 – terawatt femtosecond laser system, 2 – beam splitter, 3 – SHG, 4 – polarizing attenuator, 5 – glass wedge, 6 – photodiode, 7 – delay line, 8 – telescope, 9 – spatial filter, 10 – set of neutral light filters, 11 – micro-lens, 12 – parabolic mirror, 13 – sample, 14 – vacuum chamber, 15 – interference filters, 16 – CCD camera.

### 3. Determination of parameters of laser pump pulse

Figure 2 demonstrates temporal profile of femtosecond laser pulse in picosecond time range obtained by third-order cross-correlator with dynamic range of \( 10^8 \). Temporal profile of laser pulse as well as temporal contrast are the key features of high-power femtosecond laser pulse. Temporal contrast is the ratio of the peak power to the nanosecond “pedestal” caused by amplified spontaneous emission (ASE) in active medium of laser. As is seen from the figure, the pulse contrast at a wavelength of 1240 nm is \( 10^7 \) in the range of \( \pm 2 \text{ ps} \) and in nanosecond range as well; while at the time of \( \pm 1 \text{ ps} \) laser pulse intensity equals \( \sim 10^4 \) of the maximum; pulse duration is 100 fs FWHM.

![Figure 2](image2.png)

**Figure 2.** Third-order cross-correlation function of femtosecond laser pulse.
Using an off-axis paraboloid for laser radiation focusing imposes strict requirements on the accuracy of alignment of this element, since field aberrations of an aspheric optics exceed greatly aberrations of common spherical one. To estimate spatial distribution of energy density in the beam waist, a technique described in [27] is used. According to it, a square of radius of the crater formed on the target surface as a result of ablation is linearly related to the logarithm of laser pulse energy applied. For his purpose, iron target has been temporarily replaced with a sample of polished monocristalline silicon. When the threshold value $F_{th} \sim 200 \, \text{ml/cm}^2$ is exceeded, craters on the surface become clearly distinguishable. Figure 3 (a) demonstrates experimental dependences of the square of the minor and major axes of the ablation crater on the energy of laser pump pulse. The dependences obtained are well approximated by a family of two straight lines with different angles of inclination. This means that spatial distribution of intensity in the focal plane of the off-axis parabolic mirror is described by a combination of two Gaussian curves with parameters $r_01$ and $r_02$ in figure 3 (b) for central and peripheral parts respectively. Due to oblique incidence of laser beam and elliptic shape of craters, we have different values for X and Y axes.

This spatial distribution may result from aberrations of laser beam wavefront either due to an error in the manufacturing accuracy of the surface of the focusing element (less than $\lambda/2 \, @ \, \lambda = 633 \, \text{nm}$, according to the manufacturer), or due to distortions in the active elements of laser source itself. In practice, this leads to the fact that the central tightly focused part of the laser spot $24\times32 \, \mu\text{m}$ at $1/e^2$ level contains 95% of the entire pulse energy. This in turn means that energy of $E = 3.1 \, \text{mJ}$ is required to achieve laser intensity $I_{\text{max}} = 10^{16} \, \text{W/cm}^2$ on target surface.

![Figure 3](image_url)

**Figure 3.** (a) Dependence of the square of crater size on the incident energy of pump pulse for GaAs target. Triangles and circles – experimental values for major and minor axes respectively; lines – linear approximations. (b) Spatial distribution cross-sections of the intensity of laser radiation on the target surface.

**4. Results and Discussion**

The change in the phase of the reflected wave $\Psi_{\text{ind}}$ of the probe pulse depends on the processes running at the target surface and the intensity of the pump pulse as well; in general terms, it can be represented as a sum of several components:

$$\Psi_{\text{ind}} = \Psi_{\text{oc}} + \Psi_{\text{shift}} + \Psi_{\text{pl}},$$  

(1)

where $\Psi_{\text{oc}}$ is the phase change due to the change in the optical constants of the material due to melting process, $\Psi_{\text{shift}}$ is the phase change caused by surface movement due to expansion of the target, and $\Psi_{\text{pl}}$ is the phase change due to propagation of probe radiation through plasma layer of subcritical density. The change of $\Psi_{\text{oc}}$ due to melting of target material may have various signs: in semiconductors, for example, Si and GaAs, it is negative, while in metals it can have both positive (gold) and negative (tungsten) values.
Figure 4 (a) demonstrates an example of spatial distributions of phase changes $\Psi_{\text{ind}}$ for Si and Au targets after laser exposure with laser fluence five times above the ablation threshold value for delay times $t_{\text{delay}}$ of 200 fs and 5 ps. In experimental studies [28], it was shown that information on optical constants could be obtained on the basis of changes in amplitude and phase of the reflected wave (Fresnel equations) for time delay of 200 fs only if pump pulse intensity does not exceed $2 \cdot 10^{13}$ W/cm$^2$. At intensities above the specified value, phase change caused by layer movement of critical density, from which probe pulse is reflected, exceeds the phase change caused by alternation of optical constants of target material. The change in $\Psi_{oc}$ due to the change in optical constants upon laser action on Fe target has a positive sign [29], and due to high intensity of the pump pulse $I_{\text{max}} = 10^{16}$ W/cm$^2$, we can assume $\Psi_{oc} \ll \Psi_{\text{shift}}$.

The physical meaning of $\Psi_{\text{pl}}$ term becomes clear from figure 4 (b), where two cases of reflection of the probe pulse are presented: before the pump pulse impact (at the top) and when the plasma formed at the target surface expands as a result of laser exposure (at the bottom). In the first case, electron density profile has a stepped form corresponding to the target-vacuum interface. In the second case, probe radiation reflected from the layer of critical density propagates back through plasma layer, acquiring a positive phase shift $\Psi_{\text{pl}} > 0$. Thus, all three terms in equation (1) are assumed to have non-negative values.

As it has been shown previously [19], a key issue in determining the mechanism for creating fast electrons is the value of the hydrodynamic expansion of pre-plasma prior to the impact of the maximum of intensity of the laser pulse. The pre-plasma mentioned above can be formed either as a result of ionization of vapor cloud caused by evaporation of the target exposed to ASE of nanosecond duration at the front edge of laser pulse, or as a result of the impact of a prepulse with sufficient energy. To study the effect of both the nanosecond ASE pedestal and the prepulse on target surface, measurements of phase changes $\Psi_{\text{ind}}$ have been carried out in the range of negative delay values of the probe pulse relative to the pump pulse. The moment of $t_{\text{delay}} = 0$ designates the coincidence in time of the maxima of intensity profiles of pump and probe pulses. For time delay $t_{\text{delay}} = -2$ ps, pump pulse intensity on target surface is $\sim 10^{-7} I_{\text{max}}$ that is below evaporation threshold of target material.
Figure 5 (a) demonstrates time profile of intensity of the pump pulse on a logarithmic scale over a time range of ±3 ps relative to the intensity maximum. Figure 5 (b) shows the dependence of the phase change of the complex reflection coefficient (left axis, $\Psi_{ind}$) on time delay between pump and probe pulses in the range of $-1.7 \text{ ps} < t_{delay} < 1 \text{ ps}$. From the obtained experimental data it can be seen that for $t_{delay} = 0$ phase change is $\Psi_{ind} \approx 0.58 \pm 0.12 \text{ rad}$. The error in determining phase change is caused only by the accuracy of the Fourier method of interferogram processing, but also by the error of setting a zero delay, which is $\Delta t_{delay} \pm 60 \text{ fs}$ (determined by the duration of the laser pulse), and surface roughness. Changes of $\Psi_{ind}$ in the region of negative delays ($t_{delay} \geq -300 \text{ fs}$) correspond to laser pulse intensity $I \geq 4 \cdot 10^{13} \text{ W/cm}^2$ that exceeds ablation threshold of target material.

![Figure 5. (a) Time profile of laser pump pulse. (b) Dynamics of phase change $\Psi_{ind}$ (left axis) of the complex reflection coefficient of plasma and the magnitude of the characteristic scale inhomogeneity $L$ of plasma density (right axis).](image)

Considering the assumptions made above, the experimentally obtained phase changes $\Psi_{ind}$ of the complex reflection coefficient consist mainly of two components: 1) phase change $\Psi_{pl}$ associated with the propagation of laser probe pulse in plasma with inhomogeneous dielectric constant before and after reflection from the layer with critical density and 2) phase shift $\Psi_{shift}$ due to the displacement of plasma layer, from which the probe radiation is reflected:

$$\Psi_{ind} = \Psi_{shift} + \Psi_{pl} = 0.58 \pm 0.12 \text{ rad.} \tag{2}$$

Since each of these summands is non-negative, it can be argued that the phase change caused by pre-plasma expansion does not exceed the value of $\Psi_{ind}$ i.e. $\Psi_{shift} < 0.58 \text{ rad}$. In this case, displacement of the plasma layer with critical density of $n_{cr} = 2.9 \cdot 10^{21} \text{ cm}^{-3}$ for probe radiation at a wavelength of $\lambda = 620 \text{ nm}$ does not exceed $dz = \lambda_{probe} \Psi_{shift} / 4\pi < 29 \text{ nm}$, and the expansion velocity of the layer with critical density is about $10^3 \text{ m/s}$.

The size of characteristic inhomogeneity of plasma density $L = [(1/n_e)(dn_e/dz)]^{-1}$ [30] calculated in linear approximation and corresponding to the registered phase change of the complex reflection coefficient is shown as vertical axis on the right in figure 5 (b). As the magnitude of the change in electron density, we took the difference between the initial and critical densities (for probe pulse wavelength of $\lambda = 620 \text{ nm}$) $dn_e = n_0 - n_{cr}$ shown in figure 4 (b). Thus, experimental studies have shown that at the moment of Fe target exposure to the maximum intensity of laser pulse, plasma with a characteristic size of inhomogeneity $L < 30 \text{ nm}$ is formed on the surface.

As noted in previous study [19], optimal conditions for creating electrons with energies of about 10 keV for generating X-Ray characteristic radiation, accelerated through the vacuum heating mechanism, are observed in the case when the size of plasma inhomogeneity $L$ is small compared to the amplitude of oscillations of electrons in the electric field component of laser radiation normal to
the target. According to [18], the criterion for the action of the vacuum heating mechanism is the fulfillment of the condition \( L/\lambda < 0.1 \), which corresponds to the characteristic plasma inhomogeneity size \( L \sim 120 \text{ nm} \) for an infrared laser pulse.

5. Conclusions
The results of experimental studies of plasma formation and expansion when bulk iron target is exposed to radiation of femtosecond chromium:forsterite laser system at a wavelength of 1240 nm and pulse intensity of \( I_{\text{max}} = 10^{16} \text{ W/cm}^2 \) with a high temporal contrast of \( 10^7 \) are presented. Time-resolved method of interference microscopy was applied to study the characteristic size of inhomogeneity of pre-plasma for the first time. Assuming that the recorded changes in the phase of the probe pulse reflected from the pre-plasma layer with a critical density are determined primarily by plasma expansion, the characteristic size of plasma inhomogeneity at the moment of impact of the maximum intensity of laser pulse is \( L < 30 \text{ nm} \). Obtained results suggest that vacuum heating may be the prevailing electron acceleration mechanism in this case.

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