Ultrafast imaging interferometry at femtosecond-laser-excited surfaces

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A simple and robust setup for femtosecond time-resolved imaging interferometry of surfaces is described. The apparatus is capable of measuring both very small phase shifts ($\sim 3 \times 10^{-2}$ rad) and amplitude changes ($\sim 1\%$) with micrometer spatial resolution ($\sim 1 \mu m$). Interferograms are processed using a 2D-Fourier transform algorithm. We discuss the image formation and the physical interpretation of the measured interferograms. The technique is applied to measure transient changes of a GaAs surface irradiated with intense femtosecond laser pulses with fluences near the ablation threshold.

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1. INTRODUCTION

The interactions of intense ultrashort laser pulses with solids involve a variety of very fast transient phenomena, such as laser-induced melting, ablation of surface material, and optical breakdown in the bulk of optically transparent materials. These processes usually manifest themselves in the transient changes of the optical properties of laser-excited material. The surface phenomena can also be accompanied by transient surface deformations. The development of ultrafast time- and space-resolved optical microscopy and interferometry makes it possible to study the dynamics of these ultrafast phenomena and clarify the underlying basic physical mechanisms.

Ultrafast time-resolved optical microscopy is a combination of the pump–probe technique with optical microscopy. It allows time- and space-resolved measurements of the reflectivity of a laser-excited surface by measuring the amplitude of optical probe pulses reflected from a laser-excited surface. The spatial resolution is determined by the properties of the imaging optics, typically of the order of a micrometer. Femtosecond time-resolved microscopy has previously been used to study, for example, ultrafast laser-induced phase transitions in solids and laser ablation from solid surfaces by ultrashort laser pulses.

If one also measures the phase of reflected light by using interferometric techniques, important information about surface deformations can be obtained. Time- and space-resolved optical interference microscopy has previously been used, for example, to study laser ablation of polymer films by nanosecond pulses and picosecond-laser-induced surface acoustic waves in dielectrics. In these cases, conventional two-beam Michelson or Mach–Zehnder interferometry was used. More recently, spectral interferometry has been developed and used to investigate the expansion of femtosecond-laser-produced plasmas, femtosecond-laser-driven shocks in metals, and the dynamics of the electron gas in the bulk of laser-irradiated dielectrics.

Important characteristics of time- and space-resolved interferometric techniques are (i) the temporal resolution, (ii) the lateral spatial resolution, and (iii) the phase sensitivity. The temporal resolution is determined by the duration of the probe pulse. Lateral spatial resolution and phase sensitivity are determined by the properties of the imaging optics and also, to a large extent, by the way the interferograms are recorded and processed.

The spectral interferometry requires an imaging spectrometer and provides femtosecond time resolution together with a high phase sensitivity of approximately $\lambda/2000$. The disadvantages of this technique are (i) the lateral spatial resolution is obtained in one dimension only, and (ii) the frequency resolution of the spectrometer limits the time range to only a few picoseconds.

In a previous work with conventional two-beam interferometry, a temporal resolution at $\sim 100$ ps, a 2D-lateral spatial resolution $\sim 10 \mu m$, and a phase sensitivity $\sim 1/100$ were reported. A high phase sensitivity was achieved by taking an average of several data points obtained from independent single-shot measurements. Recently, subpicosecond temporal resolution and high phase sensitivity of $\sim \lambda/400$ was achieved, albeit at the expense of the lateral spatial resolution $\sim 40 \mu m$.

The restrictions of spectral interferometry mentioned above motivate further development of the conventional two-beam interferometry. In particular, improvements of the single-shot lateral spatial resolution and phase sensitivity are required.

We have designed a simple and robust setup for optical Michelson interferometry with a Linnik imaging configuration, which is capable of measuring both
very small phase shifts $<\lambda/200$ and amplitude changes $\sim 1\%$ with femtosecond time resolution $\sim 100$ fs and 2D-lateral spatial resolution of $\sim 1$ $\mu$m. The 2D-Fourier-transform algorithm is used to reconstruct the amplitude and phase of the interference fringes.$^{13,14}$

Our experimental technique is applied to study very small surface distortions of GaAs crystals produced by irradiation with femtoseconds laser pulses with fluences near the ablation threshold.$^{15}$ In addition to the reported preliminary results on the technique,$^{16}$ here we focus on the analysis of optical image formation, which is quite important for the optimal processing of the interferograms. A general ambiguity in the physical interpretation of the interferometric measurements is discussed in detail, and the artifacts in the reconstructed phase and amplitude maps are analyzed. The importance of possible errors introduced by the phase unwrapping procedure is emphasized.

2. EXPERIMENTAL SETUP

A schematic of the experimental setup for ultrafast Michelson interferometry with a Linnik imaging configuration is shown in Fig. 1. A $p$-polarized pump pulse (Ti:sapphire, $\tau = 100$ fs, $\lambda = 800$ nm) incident at 45° is focused on the surface with a lens ($f = 50$ cm). The excited surface area is illuminated by a weak, time-delayed probe pulse (second harmonic, $\tau = 100$ fs, $\lambda = 400$ nm) normally incident onto the surface through a high-resolution objective lens (20×, $Na = 0.3$) with a working distance of 17 mm. To achieve a spatially homogeneous surface illumination, the probe pulses are spatially filtered. The strongly divergent probe pulses, which are obtained by placing an additional lens ($f = 25$ mm) in front of the beam splitter, enable the illumination of a large surface area on the sample.

The formation of the interferogram is illustrated in Fig. 2. The interferogram is formed in the image plane (surface of the CCD chip of the camera) by the interference of an object beam with a reference beam. The object beam is the reflected probe light, shown as a cone of angle $\beta$ in Fig. 2. The reference beam is a fraction of the incident probe light passed through the reference arm of the interferometer. By suitably tilting the reference mirror, the orientation and the spacing of the interference fringes can be adjusted. In Fig. 2, $\alpha$ is the angle between the reference and the object wavefront. The ray geometry will be discussed in more detail in Section 3. The interferograms are stable in periods of several seconds, demonstrating sufficient mechanical stability of the interferometer.

Each set of measurement consists of three interferograms: (i) an initial interferogram of an unexcited surface, (ii) a transient interferogram for a given pump–probe delay time, and (iii) a final interferogram taken a few seconds after the excitation. The latter characterizes final surface modification that may have occurred. A stepping motor-controlled optical delay line allows for varying the pump–probe delay time up to a few nanoseconds with time steps of as short as 6.7 fs. The interferograms are recorded with an 8-bit-CCD camera having a sensor area of 768×512 pixels. The high-contrast interferograms with a small fringe spacing of approximately 9 pixels per fringe are well suited for the application of a Fourier-transform processing technique.

Figure 3 shows an example of an interferometric measurement for a femtosecond-laser-excited (100) surface of GaAs. The pump–probe delay time was $\Delta t = 800$ ps, and the peak laser fluence in the center of the laser excited area was $F = 0.98 F_{abl}$ ($F_{abl} = 0.2$ J/cm$^2$ for given excitation conditions). Owing to the steep angle of incidence, the bell-shaped spatial fluence distribution of the pump pulses was elongated in the plane of incidence and had typical widths (FWHM) of 140 and 100 $\mu$m in the horizontal and vertical directions, respectively.

In the transient interferogram, fringe shifts can hardly be recognized with the bare eye. On the other hand, the bright elliptical area in the center is clear evidence of an increase in the reflectivity over a laser-excited surface.$^2$

For laser fluence below the ablation threshold, the final interferogram (not shown in Fig. 3) is almost identical with the initial one, indicating that no significant changes of the optical properties and of the final surface morphology have occurred. It will be shown below that laser excitation with fluences above the ablation threshold leads to the formation of well-defined ablation craters, which can be easily detected by optical interferometry.
3. ANALYSIS AND PROCESSING OF THE INTERFEROGRAMS

The interferogram represents the spatial intensity distribution $I(x,y)$ of light on the CCD detector:

$$I(x,y) = |E_{obj}(x,y)|^2 + |E_{ref}(x,y)|^2 + 2 \text{Re}[E_{obj}(x,y)E_{ref}^*(x,y)].$$

(1)

The complex amplitudes of the interfering object and the reference waves can be written as

$$E_{obj}(x,y) = \bar{r}(x,y)A_1(x,y)e^{i\phi_1(x,y)},$$

$$E_{ref}(x,y) = A_2(x,y)e^{i\phi_2(x,y)}.$$  (2)

The complex reflection coefficient of the investigated surface,

$$\bar{r}(x,y) = r(x,y)e^{i\Psi(x,y)},$$

(3)

represents the main quantity of interest. In each interferometric measurement an initial and a transient interferogram are recorded:

$$I_{in} = r_{in}^2A_1^2 + A_2^2 + 4r_{in}A_1A_2 \cos(\phi_1 - \phi_2 + \Psi_{in}) \quad \text{(initial)},$$

(4)

$$I_{tr} = r_{tr}^2A_1^2 + A_2^2 + 4r_{tr}A_1A_2 \cos(\phi_1 - \phi_2 + \Psi_{tr}) \quad \text{(trans.)}. $$

(5)

From the interferograms (4) and (5) we reconstruct the actual laser-induced changes in the amplitude and phase of the object wave as compared with an unexcited surface:

$$\Psi_{in}(x,y) = \Psi_{tr}(x,y) - \Psi_{in}(x,y) \quad \text{(transient phase shift)},$$

(6)

$$r_{in}(x,y) = r_{tr}(x,y)/r_{in}(x,y) \quad \text{(transient ampl. change)}. $$

(7)

Note that expressions (6) and (7) provide the laser-induced changes in the complex reflection coefficient of the surface.

In the case of a properly aligned microinterferometer, the interferograms consist of many parallel vertical and almost equidistant interference fringes. The spatial dependence of the phase of the interference fringes is then linear:

$$\phi_1(x,y) - \phi_2(x,y) = 2\pi f_0 x + \text{const.}$$

(8)

The carrier frequency of the interference fringes, $f_0$, is proportional to the angle $\alpha$ between the object and reference wavefronts (see Fig. 2).

Since the interferometer is illuminated by spatially filtered pulses, the intensity distribution for both probe and reference pulses is spatially homogeneous:

$$A_1(x,y) = \text{const}, \quad A_2(x,y) = \text{const}. $$

(9)

Using approximations (8) and (9), the general expression for the interferograms (4) and (5) can be simplified as follows:

$$I(x,y) = [1 + r^2(x,y)] + 2r(x,y)\cos[2\pi f_0 x + \Psi(x,y)],$$

(10)

where equal amplitudes $A_1 = A_2 = 1$ are assumed. The analysis of a simplified and approximate expression (10) will be useful for the understanding of the interferogram formation in the Linnik microinterferometer and the physically relevant aspects of the algorithm used for the reconstruction of the phase of the interference fringes in a case of high carrier frequency. An overview on Fourier-transform techniques is given in Ref. 19. To illustrate the different steps of the procedure, we apply it to the transient interferogram of Fig. 3(b).

First, an interferogram is multiplied by a so-called window, which is a function that decreases from a maximum value in the center to a minimum value at the boundaries of the interferogram. We apply a Hamming window, widely used for interferogram processing. The purpose of “windowing” is to reduce the effects of the sharp boundaries in the 2D-Fourier transform. Figure 4(a) shows the distribution obtained by applying a Hamming window to the recorded interferogram [Fig. 3(b)].

The second step is to perform the 2D Fourier transform of the windowed interferogram of Fig. 4(a). The absolute square of the Fourier transform is shown in Fig. 4(b). It is plotted on a logarithmic scale, since the magnitude of the 2D Fourier transform varies over many orders of magnitude.

Because of the sampling of the interferogram during the recording by a CCD chip, the range of spatial frequencies is bounded by Nyquist frequencies $f_N = 1/(2\Delta)$, where $\Delta = \Delta_{CCD}/M = 300$ nm is the pixel size of an interferogram and $\Delta_{CCD} = 9 \, \mu m$ and $M = 30$ are the pixel sizes of the CCD camera and the optical magnification, respectively.

In Fig. 4(b), three distinct peaks can be recognized in the Fourier plane. The central peak at zero frequency (dc peak) corresponds to the term $[1 + r^2(x,y)]$ in Eq. (10).
The amplitude less than $10^{-2}$ compared with the ac peaks. These structures represent harmonics caused by the nonlinear response of the CCD camera.20,21

The basic idea of the Fourier-transform technique is to isolate one of the ac peaks in the frequency domain by spectral filtering. The inverse Fourier transform of a filtered spectra then provides the phase and amplitude of the interference fringes. Since the location and spread of useful signal components in the Fourier plane is usually unknown, the choice of a spectral filter with suitable size and shape is not a trivial problem.

To understand which parts of a Fourier plane contain the desired information about the object wave, let us consider a well-known process of the image formation in a microscope objective with a certain numerical aperture.22–24 Only a finite cone of rays coming from the object is collected by the microscope objective and contributes to the formation of the image. The rest is cut off by the aperture. Therefore an upper limit of the spatial frequencies of the object wave $r(x,y)\exp[i\Psi(x,y)]$ is given by the relation

$$f < f_{\text{max}} = \frac{\sin \alpha}{\sin \beta} \cdot \frac{\lambda}{N A},$$

where $\lambda$ is the wavelength of the probe pulses and NA is the numerical aperture. In our experiments, the maximum frequency is $f_{\text{max}} = 0.45f_N$.

These considerations are important for the Fourier transform of Eq. (10), which can be analyzed using a convolution theorem: The spectral components of the two ac peaks must be localized within the black circles of radius $f_{\text{max}}$ centered at frequencies $\pm f_0$ [black dashed circles in Fig. 4(b)], whereas the dc-peak components are localized within the circle of the radius $2f_{\text{max}}$ [white dashed circle in Fig. 4(b)]. This result allows for a simple geometrical interpretation in terms of the angles $\alpha$ and $\beta$ in Fig. 2:

$$\frac{f_0}{f_{\text{max}}} = \frac{\sin \alpha}{\sin \beta} < 1.$$  (12)

This demonstrates the importance of the proper adjustment of the interferometer but also reveals an important limitation of the Linnik microinterferometer. Indeed, it can be easily inferred from Fig. 2 that the maximal value of the angle $\alpha$ in the Linnik interferometer is limited by $\beta$, or, which is just the same that the career frequency $f_0$ cannot exceed the cutoff frequency $f_{\text{max}}$. For the presented interferograms, both angles are very small: $\alpha = 0.5 \cdot 10^{-2}$ rad and $\beta = 10^{-2}$ rad.

Summarizing the results of the above analysis, the main conclusion is the components of the ac peaks [localized within the black dashed circles in Fig. 4(b)] cannot be fully separated from the central dc peak [localized within the white dashed circle in Fig. 4(b)]. This fundamental limitation is determined by the interferogram formation in a Linnik microinterferometer and makes it impossible to clearly separate the spectral components describing the amplitude and the phase of interference fringes in a Fourier plane.

Nevertheless, under certain conditions, one of the ac peaks in a Fourier plane (we have chosen the right one) can be reasonably isolated by means of spectral filtration in such a way that the important information about the amplitude and phase of the interference fringes is not lost. We have used an elliptical Hamming filter with the following properties: its vertical size is chosen to be equal to the cutoff frequency of the microscope objective to maintain the best possible spatial resolution in the $y$ direction, whereas its horizontal size is four times smaller.

![Fig. 4](image)

(a) Transient interferogram [Fig. 3(b)] multiplied by a Hamming window, (b) 2D Fourier transform of (b), and (c) isolated right ac peak of (b).

![Fig. 5](image)

(a) Reconstructed transient phase shift $\Psi_{\text{ind}}(x,y)$ and (b) amplitude change $r_{\text{ind}}(x,y)$. Plot (c) represents the transient phase and amplitude profiles along the vertical cross sections in (a) and (b). Frame size: $180 \mu m \times 130 \mu m$, $\Delta t = 800$ ps.
in order to obtain a reasonable separation of the ac-signal components from the central dc peak [Fig. 4(c)].

The angle of the inverse Fourier transform of the filtered Fourier spectra equals the total phase of the transient interferogram \( \phi_1(x,y) - \phi_0(x,y) - \Psi_{\text{ind}}(x,y) \), and its amplitude is equal to \( 2r_{\text{ind}}(x,y)A_1(x,y)A_0(x,y) \). The initial interferogram is subjected to the same processing as the transient one, and the laser-induced phase shift \( \Psi_{\text{ind}} \) and amplitude change \( r_{\text{ind}} \) are calculated and are presented in Fig. 5. The phase shift is unwrapped (i.e., the \( 2\pi \) discontinuities removed) using the simple unwrapping algorithm of Takeda et al.\(^\text{17} \)

In the reconstructed phase and amplitude maps, the spatial resolution is determined by the size and shape of the response function of the spectral filter, given by the Fourier transform of the filter function. The response function is plotted in the right upper corner of Figs. 5(a) and 5(b). Because we use an elliptically shaped Hamming filter [see Fig. 3(b)], the corresponding response function is also elliptical, and the lateral spatial resolution in the horizontal and vertical directions \( x \) and \( y \) is different:

\[
\Delta x = 6.0 \mu\text{m}, \quad \Delta y = 1.5 \mu\text{m}.
\]  

(13)

The spatial resolution in the \( x \) direction is significantly reduced below the Rayleigh resolution limit \( 0.6\lambda/\text{NA} = 0.8 \mu\text{m} \) of the microscope objective. This is necessary to improve the suppression of the central spectral components (dc component). The bell-shaped profile of the Hamming filter helps to minimize the errors due to the boundary effects of the Fourier transform.\(^\text{13} \) Because the spatial resolution in the \( y \) direction is higher, we analyze vertical profiles of the phase and amplitude maps. The examples presented in Fig. 5(c) will be discussed in detail in Section 4.

As a measure of the experimental accuracy, we use the root mean square (rms) of the phase and the amplitude measured over an unperturbed area of the crystal surface:

\[
\delta\Psi_{\text{ind}}^{\text{(rms)}} = \pi/100, \quad \delta A_{\text{ind}}^{\text{(rms)}} = 0.01
\]  

(14)

The phase accuracy of \( \pi/100 \) indicates that surface deformations as small as 1 nm can be detected with our apparatus.

To conclude this section we mention one technical aspect of interferogram processing. A significantly improved performance of the Fourier-transform technique could be achieved by the application of the so-called correction for the continuum.\(^\text{14,25} \) The idea is first to estimate somehow the background illumination (dc peak), which can be used to correct the interferogram. The Fourier spectra of such a corrected interferogram does not contain a dc peak, which leads to a better isolation of the desired signal components of the ac peaks. Unfortunately this approach is limited to the situation when the background illumination is always the same. The background illumination of our transient interferograms is influenced by transient reflectivity changes, and such a simple correction procedure cannot be applied.

4. PHYSICAL INTERPRETATION OF INTERFEROMETRIC MEASUREMENTS

In Section 3, we reconstructed the phase shifts and amplitude changes of the light wave upon the reflection from the laser-irradiated GaAs surface, 800 ps after the laser excitation presented in Fig. 5. As already mentioned in the Introduction, the optical properties of the surface might change owing to (i) changes of the optical constants upon laser excitation and (ii) surface deformations. Without the loss of generality, the spatial variation of optical properties along the single spatial coordinate \( x \) is considered. Jumping ahead with physical interpretation, we state that the laser-excited material will be molten and the central part of the melt expands toward the vacuum and, at some moment in time, gives rise to transient surface deformation \( d(y) \). The complex index of refraction \( n(y) = n(y) - i\kappa(y) \) is also varying as a function of \( y \). In our example, the index is different for solid and liquid phases. The reflection of a plane monochromatic electromagnetic wave \( \text{exp}(i\omega t + ikz) \) normally incident on such a deformed surface with spatially varying complex index of refraction can be formally described by the complex reflection coefficient:

\[
P(x) = r(y)e^{i(d(y) + 2kd(y))},
\]  

(15)

with \( k = (2\pi/\lambda) \), and the quantities \( r \) and \( \phi \) are given by a well-known Fresnel formula for a reflection coefficient \( F_{Fr} \) of an undeformed surface,

\[
F_{Fr}(y) = r(y)e^{i\phi(y)} = \frac{1 - \tilde{n}(y)}{1 + \tilde{n}(y)}.
\]  

(16)

The formulas (15) and (16) are derived using the convention of an \( e^{int} \) process and the Fresnel convention for the sign of \( F_{Fr}(y) \), which represent one of the four possible sets of conventions.\(^\text{26} \) The choice of conventions is obviously irrelevant for physical interpretation.

Fig. 6. Temporal evolution of transient phase shift \( \Psi_{\text{ind}} \) and amplitude change \( r_{\text{ind}} \) at points B and C in Fig. 5(c).
The interferometric measurements provide information about the amplitude \( r(y) \) of the reflected object wave and its total phase \( [\phi(y)+2kd(y)] \). Whereas the amplitude depends only on the complex index of refraction, the phase also contains the term proportional to surface deformation. The two measured quantities are obviously not enough to determine the three unknown parameters, i.e., \( r(y), \phi(y), d(y) \) in Eq. (15). Therefore some additional information or assumptions are inevitably required to provide the physical interpretation of the experimental data. In Subsection 4.A, it will be shown which kind of information can be useful to interpret interferometric data for the subthreshold excitation of the GaAs surface shown in Fig. 5.

A. Optical Constants and Deformations of a Laser-Molten GaAs Surface

Let us focus on the spatial phase and amplitude profiles of Fig. 5(c). Starting from the reference values of an unexcited solid (point A), the amplitude rapidly increases by 13% whereas the phase decreases by \(-0.08 \pi\) (point B). In the vicinity of point B, the amplitude and phase do not change as a function of the coordinate forming a plateau. In the center (point C), the phase reaches the maximum of \(-0.4 \pi\), and the reflectivity slightly decreases.

The formation of a plateau in the amplitude of reflected light (in the vicinity of point B) represents the area of high-constant surface reflectivity and is consistent with the previous observations provided by time-resolved microscopy.\(^2\) It can be physically interpreted as a result of the laser-induced melting of semiconductors, whereas the thickness of the molten layer is larger than the skin depth for a probe wavelength.

Our time-resolved interferometric measurements (Fig. 6) show that at point B both the amplitude and phase approach constant values very fast, within approximately 1 ps, indicating an ultrafast nonthermal melting process\(^2\) and do not change significantly until a few tens of nanoseconds when the liquid semiconductor cools down and re-solidifies. The time needed for transient surface deformations to develop is governed by the speed of sound and is much larger than 1 ps. Therefore we conclude that point B in Fig. 5(c) describes an undeformed laser-molten GaAs layer with a thickness larger than the skin depth of the probe light. The observed decrease of the phase is thus associated mainly with the changes of the optical constants according to the Fresnel formula [Eq. (16)].

To calculate the optical constants of liquid GaAs, it should be noted that out-interferometric amplitude and phase measurements are relative with respect to an unexcited solid. A substitution of the optical constants of unexcited GaAs, \( n_{solid}=4.373-2.146i \) at \( \lambda=400 \text{ nm} \),\(^{27}\) in the Fresnel formula [Eq. (16)] provides its complex reflection coefficient \( r_A=0.69 e^{0.94 \pi} \). Using the measured phase shift and amplitude change between points A and B, the complex reflection coefficient \( r_B=0.78 e^{0.96 \pi} \) for liquid GaAs is obtained. By inverting the Fresnel formula [Eq. (16)], we immediately get the complex index of refraction of liquid GaAs,

\[
\tilde{n}_{liquid} = 2.0 - 3.4i. \tag{17}
\]

This result can be interpreted within the frame of a simple Drude model for the optical constants of a metal,

\[
\tilde{n}_{Dr} = \sqrt{1 - \frac{\omega_p^2}{\omega^2 - i/\tau_e}}, \tag{18}
\]

where \( \omega_p=\sqrt{\epsilon_0 N_e m^*} \) is the plasma frequency. The two most important and unknown parameters in this model are collision time \( \tau_e \) and electron concentration \( N \). The experimentally determined complex index of refraction [Eq. (17)] for liquid GaAs can be fitted by a Drude model with the following parameters: \( m^*=m_e, \tau_e=0.134 \text{ fs} \) and \( N=2.1 \times 10^{23} \text{ cm}^{-3} \). These values are in good agreement with previously reported estimations from experimental reflectivity measurements\(^{28}\) \((m^*=m_e, \tau_e=0.125 \text{ fs}, N=1.95 \times 10^{23} \text{ cm}^{-3})\) and theoretical estimations based on a molecular dynamics simulation of liquid GaAs (Ref. 29) \((m^*=m_e, \tau_e=0.15 \text{ fs}, N=1.9 \times 10^{23} \text{ cm}^{-3})\).

The interpretation of the phase and amplitude changes observed in the center of laser-irradiated area (point C) is not straightforward. The analysis of temporal evolution (Fig. 6) shows that the phase shift at point C evolves in time on a much larger time scale of the order of 1 ns, on which transient surface deformations can take place. However, the phase shift can also be affected by some slow dynamics of the optical constants.

![Fig. 7. (a) Physical interpretation of interferometric measurements. Complex reflectivity vectors for three points in Fig. 5(c): A, unexcited GaAs; B, liquid GaAs; C, expanded surface. The dark region contains all possible complex reflection coefficients of an undeformed surface. (b) Surface plot representation of the transient phase in Fig. 5(a).](image-url)
To estimate possible contributions of surface deformations to the phase shift, we shall analyze the general formula [Eq. (15)]. It is very convenient to present an arbitrary complex reflection coefficient [Eq. (15)] by a vector on a complex reflectivity diagram, Fig. 7(a). The dark half-circle in Fig. 7(a) contains all possible complex reflection coefficients for an undeformed surface given by Eq. (16) for all possible optical constants $n > 0, k > 0$. Thus the reflection coefficients [Eq. (16)] of any undeformed surface are obliged to lie inside the dark area.

The complex reflection coefficients $\tilde{r}_A, \tilde{r}_B, \tilde{r}_C$ in Fig. 7(a) represent the corresponding points in Fig. 5(c). The complex reflection coefficients $\tilde{r}_A$ and $\tilde{r}_B$ lie within the dark area, which is in agreement with our previous conclusions; they describe undeformed solid and liquid GaAs surfaces, respectively. Vector $\tilde{r}_C = 0.74e^{1.33\pi}$ lies far outside of the dark region. Therefore it cannot be explained by a pure change of the optical constants, but represents transient surface excursion. Assuming that the phase difference of $\delta\phi = 0.47\pi$ between $\tilde{r}_B$ and $\tilde{r}_C$ is mostly induced by surface excursion, we get an estimate of its amplitude:

$$d = (\lambda/4\pi)\delta\phi = 47\text{ nm}.$$  \hspace{1cm} (19)

It is impossible to obtain the exact value for the amplitude of the surface deformation. However, since the reflectivity value at point C lies in between the values for liquid and solid GaAs, it is reasonable to assume that the phase shift due to changes in the optical constants does not exceed $\delta\phi = 0.08\pi$. The latter value provides a plausible empirical measure for the accuracy of $\sim 10$ nm of the estimate [Eq. (19)].

Summarizing this subsection, the physical meaning of interferometrically measured phase can be best visualized in a surface plot of transient phase, Fig. 7(b). Whereas a dark oval area (negative phase shift, point B) represents the laser-molten GaAs surface, the bright hill in the center (positive phase shift, point C) represents the surface deformation. The results of time-resolved measurements presented in Fig. 6 show that transient surface excursion at point C is fully reversible.

![Image](image.png)

Fig. 8. (a) Transient interferogram at ablating GaAs surface ($F = 1.3F_{abl}, \Delta t = 1.8$ ns), (b) 2D Fourier transform of the windowed transient interferogram, (c) isolated right ac peak, (d) horizontally unwrapped phase $\Psi_{ind}$, (e) vertically unwrapped phase $\Psi_{ind}$, (f) cross sections of phase maps (d) and (e), and (g) inconsistent phase values.

It immediately follows from the above analysis of the optical constants that large phase shifts of the order of an optical wavelength or larger are mostly induced by surface deformations. They can generate complex reflectivity vectors that lie inside the dark area in Fig. 7(a) after several rotations, which is a trivial case not discussed above.

### B. Example of an Interferometric Measurement at the Ablating GaAs Surface

So far we have illustrated and discussed the details of the interferometric technique by using only one example, i.e., the transient deformation of a GaAs surface excited slightly below the ablation threshold. In this subsection, we shall present and discuss in detail the second example of transient dynamics of the ablating GaAs surface, which aims to reveal some problems and drawbacks of the interferometric technique.

Figure 8 shows an example of transient interferometric measurement at an ablating GaAs surface excited 30% above the ablation threshold for the pump-probe delay time of 1.8 ns. A big shift and strong modulation of contrast of interference fringes within the laser-excited area are quite apparent in the transient interferogram, Fig. 8(a). The Fourier transform of a windowed transient interferogram is presented in Fig. 8(b): a significant spread of the signal component of the ac peaks is quite apparent. Because of this spread, some important signal components, primarily in the horizontal direction, are inevitably lost in the filtered Fourier transform, Fig. 8(c). This leads to the fact that the reconstructed phase cannot be correctly unwrapped. The latter statement is illustrated by Figs. 8(d) and 8(e), which represent the result of the unwrapping of the reconstructed phase map $\Psi_{ind}(x,y)$ line by line in horizontal and vertical directions, respectively. The two maps are obviously different and contain a lot of stripes. At the location of the stripes, phase experiences multiples-of-$2\pi$ jumps, which is a clear manifestation of the unwrapping problems.

The problems with unwrapping are best visualized in Fig. 8(f), where the spatial phase profiles along the white
arrows of maps in Figs. 8(d) and 8(e) are plotted. A vertical phase cross section [obtained from the phase map of Fig. 8(e)] possesses a bell-shaped profile. A big maximum phase shift in the center of the laser-excited area is \(\sim 5.5\pi\) and is mostly induced by a substantial geometric displacement of the ablating surface of \(\sim 550\) nm. For this reason, the phase is given in nanometers, and spatial phase profiles can be associated with the actual relief of the ablating surface. A horizontal phase cross section [obtained from the phase map of Fig. 8(d)] exhibits the two sharp negative jumps. The forthcoming discussion aims to demonstrate that the vertical phase profile is correct and represents the actual surface deformation, but the horizontal one contains artifacts. As already mentioned above, we have a much better spatial resolution in the vertical direction determined by a strongly asymmetric shape of the spectral filter. If the size of the spectral filter is reduced to that in the horizontal direction, then the unwrapped vertical phase profile also possesses two negative jumps. This is a clear indication that the observed artifacts in the reconstructed phase are due to the loss of signal components during spectral filtering. This phenomenon is called aliasing or energy leakage.\(^{30,31}\)

There exists a very useful interferometric trick, which allows us to find the so-called inconsistent phase values that cannot be unwrapped by any unwrapping algorithm in principle. The idea is to compare the phase value \(p_{x,y}\) at an arbitrary point \(x,y\) of the phase map with phase values at three neighboring points. An artificial sequence of five phase values \(p_1=p_{x,y}, p_2=p_{x+1,y}, p_3=p_{x+1,y+1}, p_4=p_{x,y+1}, p_5=p_{x,y}\) is unwrapped. If \(p_5\) differs from \(p_1\) after unwrapping, all four pixels are marked as containing inconsistent phase values. These inconsistent phase values are often referred to as aliasing-induced dislocations.\(^{31}\)

Figure 8(g) shows the positions of aliasing-induced dislocations for the reconstructed phase discussed above. There are relatively few dislocations all of them lying on an elliptical ring around the center of the laser excited area, where the spatial gradient of the phase (surface position) is maximal. By using the knowledge of where the dislocations come from and exploiting the elliptic symmetry of laser excitation, we have developed our own unwrapping algorithm, which will be discussed in Subsection 4.C.

Before starting to tackle the problem of unwrapping phase maps containing inconsistent values, we shall present the last example of interferometric measurement for the final surface morphology of a GaAs surface after laser ablation. The reason for doing this first will become clear later.

Figure 9 illustrates the processing of the final interferogram of a GaAs surface after laser excitation with the peak fluence 30% above the ablation threshold: final interferogram [Fig. 9(a)], the Fourier spectra of a windowed final interferogram [Fig. 9(b)], filtered Fourier spectra [Fig. 9(c)], surface plot of the reconstructed phase shift [Fig. 9(d)], and an automatically determined position of the boundary of the ablation crater [Fig. 9(e), to be explained later]. The final interferogram Fig. 9(a) contains an elliptical ring at which interference fringes experience a jump. This jump represents the boundary of the ablation crater and is induced by the permanent removal of a macroscopic amount of the material from a solid surface as the result of laser ablation.\(^3\) A detailed microscopic investigation of the final surface morphology of a GaAs surface using atomic force and electron microscopy\(^{32}\) showed that the boundary of the ablation crater is extremely narrow with a width around 100 nm. This should obviously lead to the cutoff of some signal components by the objective aperture. The signatures of such cutoff can indeed be recognized in the Fourier spectra [Fig. 9(b)] in the form of the two dark circular areas with the diameter \(2f_{\text{max}}\) around two ac peaks. These circular areas are displaced not only horizontally, as expected from our simple analysis [see Fig. 4(b)], but also vertically. An extended routine analysis (not discussed here) shows that additional vertical displacement of the two circles in the Fourier plane can be explained by a slight misalignment of the interferometer, in which reference and object beams are shifted vertically with respect to the optical axes of the microscope objectives. Since the spacing and vertical orientation of interference fringes remain unchanged for this type of misalignment, it can hardly be recognized by the visual inspection of interferograms.

It was demonstrated earlier that the reconstructed phase surface [Fig. 9(d)] represents the actual relief of the final ablation crater on GaAs (Ref. 32) and, for this reason, is again recalculated in nanometers of surface displacement. The ablation crater has a perfect elliptical shape, possesses extremely steep walls, and has a depth of 40 nm, which is weakly dependent on the applied laser
fluence. Because of these steep walls, it is technically easy to determine the position of the crater boundary, say at half the crater depth, and fit it with an ellipse. The result of this procedure is presented in Fig. 9(e). The knowledge of the position and size of the ablation crater provides precise values for the position and the energy of the pump pulse used to excite the sample, which is important for fully automatic processing of interferometric data.

C. Unwrapping Algorithm

Provided with the exact position of the excitation pulse on the sample, we return to the problem of unwrapping phase maps containing dislocations [see Figs. 8(d) and 8(e)].

According to our two-step algorithm, the transient phase map is first unwrapped line by line along elliptical paths starting from the center of the laser excited area [Fig. 10(a)]. Unwrapping fails at only a few elliptical paths containing dislocations [see Fig. 8(g)], which can be best recognized in the vertical and horizontal cross sections of the phase, Fig. 10(b); a few sudden phase jumps can clearly be seen in both profiles. As the last stage of unwrapping, we decided to use the so-called band limit approach (global feedback): “the approach is analogous to a human observer adding at arbitrary phase step function to the wrapped data until the result appears smooth and continuous for the eye.” The elliptically unwrapped phase [Fig. 10(a)] is subjected to the 2D-Fourier transformation. The Fourier transform contains high-frequency components induced by a few 2π-phase jumps on elliptical rings with phase dislocations. High-frequency components in Fourier spectra are removed by spectral filtering. The inverse Fourier transform of such filtered spectra generates a smoothed phase. The smoothed phase is compared with the original one for each pixel: If the phase difference is larger than π, then a multiple of 2π is added to the original phase to minimize the difference, otherwise the original phase remains unchanged. This procedure is repeated several times to generate the unwrapped phase map, presented in Fig. 10(c). Note that to generate phase map (c) from phase map (a), no image processing has been applied except for adding multiples of 2π at a few pixels. Vertical and horizontal profiles of the phase [Fig. 10(d)] and phase surface [Fig. 10(e)] demonstrate a good performance of the described unwrapping algorithm.

D. Movement of the Ablating GaAs Surface

The results of time-resolved interferometric measurements for excitation above the ablation threshold are presented in Fig. 11, where the position of the ablating surface in the center of the laser-excited area (fluence \( F = 1.25 F_{\text{abl}} \)) is plotted as a function of time. The data delivered by interferometry denoted as row data in Fig. 11 are obtained from unwrapped phase maps for different pump–probe delay times. The surface motion is expected to be continuous in time, most likely with a constant velocity for excitation fluences above the ablation threshold. The temporal dependence of the phase shows an unexpected discontinuity at pump–probe delay times around 2 ns. However, being artificially raised by 200 nm = 2π (corrected data in Fig. 11), several points for large delay times perfectly extrapolate the data for shorter delay times. The corrected data in Fig. 11 represent the expansion of an ablating material with a constant velocity around 270 m/s, in agreement with the results of the previous investigation.

The explanation for the observed artificial phase jumps directly follows from the above discussion of unwrapping problems in the previous sections. Phase maps for different pump–probe delay times (not shown) show that big gradients of surface position develop with time. Even a high spatial resolution in the vertical direction becomes insufficient to resolve strong spatial phase variations. Only the inspection of temporal evolution makes it evident where the missing multiples of 2π should be added.

E. Are the Reconstructed Maps of the Surface Reflectivity Correct?

To check whether the reflectivity maps reconstructed from the interferogram are correct, we have measured the actual laser-induced changes of the surface reflectivity by conventional time-resolved microscopy. Figure 12 con-

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Fig. 10. (a) Elliptically unwrapped transient phase of Fig. 8(d), (b) vertical and horizontal phase cross sections of (a), (c) result of unwrapping of phase map (a) using the band-limit approach, (d) vertical and horizontal phase cross sections of (c), and (e) surface plot representation of transient phase (c).
pares the directly measured surface reflectivity with that reconstructed form of the transient interferogram of Fig. 8(a). The pumping conditions and pump–probe delay time in both cases were the same. The difference between the directly measured and reconstructed reflectivity maps is quite apparent; the reconstructed map contains many more dark regions corresponding to a lower surface reflectivity.

To explain this difference, we shall again refer to the topical problem of aliasing or energy leakage. According to Parseval’s theorem, the integrals of the squared signal in coordinate space and Fourier space are equal. Since some frequency components of the signal are lost by spectral filtering, the reconstructed reflectivity should be smaller than the real one. To verify this conclusion, we have compared the directly measured reflectivity profiles with those obtained using spectral filters of different shapes and sizes for several interferometric data sets. Not only the size but also the shape of the spectral filter are found to be important. In the case of strong spatial reflectivity modulations or phase gradients, the reconstructed reflectivity values are always somewhat lower as compared to the direct reflectivity measurements. Correct values of surface reflectivity are obtained only in the case of smooth spatial dependencies of reflectivity and phase.

Not only the reconstructed but also the directly measured reflectivity maps can contain artifacts. This is the case, for example, if the tilt angle of the ablating surface with respect to the unexcited surface is comparable with the opening angle of the microscope objective (~17.5°). Then some rays coming from the microscope objective do not enter the objective aperture after reflection from strongly tilted surface areas, leading to the formation of conspicuous dark rings. In this case, the cutoff circles by an aperture of the objective can be recognized in 2D Fourier spectra of the directly measured reflectivity maps.

5. CONCLUSIONS AND PERSPECTIVES

In this paper, we have presented and discussed different aspects of ultrafast time-resolved imaging interferometry. Particular attention was paid to the optical image formation in the Linnik microinterferometer, which is essential for the correct application of the 2D-Fourier-transform algorithm for phase and amplitude reconstruction. Physical interpretation of interferometric measurements has been discussed using the experimental data for a femtosecond-laser-excited GaAs surface below and above the ablation threshold. A rather general analysis of the complex reflection coefficients of laser-excited surfaces makes it possible to distinguish phase contributions induced by changes in the optical constants and surface deformations. Surface deformations with amplitudes of a few tens of nanometers can be clearly identified. For short pump–probe delay times in the range of a few picoseconds, when transient surface deformations have not yet developed, interferometric measurements represent a precise tool for measuring transient laser-induced changes in the optical constants.

Some technical questions were addressed to phase unwrapping algorithms and their failure in certain cases. Analysis of both spatial and temporal variations of the phase allows for the identification of possible artifacts of the interferometric measurements. Phase measurements are found to be quite reliable, whereas major phase errors represent multiples-of-2π jumps, which can be easily recognized and removed both manually and by using fully automatic algorithms. In contrast, in certain cases the reflectivity maps reconstructed from the interferograms exhibit artifacts cannot be eliminated in principle. A straightforward explanation for these observations is provided by considering imaging with a microscope objective of finite numerical aperture and application of additional spectral filtering during interferogram processing.

Beyond the experiments at laser-excited surfaces, time-resolved interferometry can also be applied to investigate ionization mechanisms in the bulk of optically transparent materials using imaging Mach–Zehnder interferometry (in transmission). It provides similar results as compared to the spectral interferometry. One of the promising applications for interferometry in transmission is the investigation of femtosecond-laser-induced optical
breakdown and shock waves in water, which were previously investigated by ultrafast microscopy.\(^\text{34}\)

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