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Femtosecond plasmon interferometer

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ABSTRACT

We have realized a plasmonic interferometer formed by a nanoslit and a nanogroove in a single-crystal gold film. The possibility of measuring laser pulses of ultimately short durations, corresponding to two periods of a light wave (6 fs pulse duration), has been demonstrated using this interferometer.

1. Introduction

Localization of light in space (on a nanometer scale) and time (on a femtosecond scale) is an important fundamental problem, which has applications in all-optical high-speed nano-optics components [1,2], nanoplasmonics [3], nanophotonics [4], and quantum devices for information processing [5,6]. Nanolocalization of light in space can only be realized by using spatially localized plasmonic waves excited in nanostructures [7]. The use of femtosecond laser radiation for excitation of the plasmonic waves makes it possible to realize nanometer and femtosecond localization of light [8].

Among a large variety of femtosecond laser sources, those that can emit light of an ultimately short duration, shorter than the relaxation time of corresponding plasmonic oscillations, are of special interest in the field of nanoplasmonics. In this case, it becomes possible to investigate ultrafast processes related to the coherent dynamics of plasmonic oscillations, i.e., to perform investigations in the field of coherent nanoplasmonics.

The minimum achievable duration of a laser radiation pulse is roughly equal to two oscillation periods of the light wave [9]. In the visible spectral range, two-cycle laser pulse radiation has a broad emission spectrum, which extends from 650 nm to 1 μm. Such a large width of the emission spectrum requires dispersion control of the medium over the entire propagation path from the laser to the sample. This imposes strong restrictions on the use of such laser pulses in experiments in nanoplasmonics.

The group velocity dispersion and laser pulse duration can be measured using a commercially available ultrashort pulse measurement technique, e.g., a spectral phase interferometer for direct electric field reconstruction (SPIDER) system. During the measurements, both the nonlinear crystal of the SPIDER system and sample with plasmonic nanostructures should be arranged at the same distance from the laser. This should be done with high precision to guarantee that the laser pulse duration for the sample is the same as that measured by the SPIDER system. However, it is not always possible to realize equal corresponding optical paths of the laser radiation (from a laser to the nonlinear crystal of the SPIDER system and from the laser to the sample with plasmonic nanostructures). As a rule, to realize interaction with nanostructures, the wave front of the laser radiation is specially constructed using various optical elements (lenses, objectives, phase plates, dielectric mirrors, etc.). These elements strongly affect the laser pulse and lead to a dephasing effect on its spectral components. As a consequence, the duration of the laser radiation pulse can be significantly and uncontrollably changed. Therefore, commercially available systems for measuring the pulse duration (SPIDER, etc.) cannot be used to monitor the duration of a pulse that directly interacts with the plasmonic nanostructure.

In experiments on nanoplasmonics, it would be ideal to measure the duration of a laser pulse directly in the specimen plane of the microscope [10]. This was the objective of the present work, in which we present results of our experimental investigations on realization of a plasmonic tilted “slit-groove” (TSG) interferometer

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[11], which was prepared on a gold film, and the duration of a pulse of femtosecond laser radiation was determined using this interferometer. The possibility of monitoring the pulse duration in the interval from 6–50 fs is demonstrated.

2. Plasmon interferometer

Fig. 1a shows a schematic image of a TSG interferometer prepared on a gold film, including a slit and a groove tilted at a certain angle relative to the slit. When measuring the pulse duration, the interferometer was illuminated by laser radiation perpendicular to the film plane. The scattering of the laser light by the groove leads to an excitation of a surface plasmonic wave with wave vector k_{sp}, which is perpendicular to the groove [12]. The plasmonic wave (SPP on the figure) propagating from the groove in the direction towards the slit is partially scattered by the slit, and it creates a near field in the region of the nanoslit with amplitude E_2. Owing to the angle α between the slit and the groove, the distance traveled by the plasmonic wave from the groove to the slit varies linearly along the slit axis x as: $d(x) = d_0 + x\sin(\alpha)$; here, d_0 is the minimal distance between the slit and groove. This leads to the dependence of the field amplitude on the coordinate x: $E_2(x) \sim \exp(-k''_{sp}d(x))$ and to a similar dependence of its phase: $\Phi(x) = k'_{sp}d(x) + \phi_0$. The angle between the slit and groove causes interference on the slit of the two near fields; one originating from the laser light illuminating the slit and the second from the plasmonic wave propagating from the groove:

$$I(x) = E_1(x)^2 + E_2(x)^2 + 2E_1(x)E_2(x)\cos(\Phi(x)), \quad (1)$$

where $E_1(x)$ is the amplitude of the near field originating from the laser light in the slit. The interference term in expression (1) has a spatial period, which is defined by the expression:

$$T_x = 2\pi/(k'_{sp}\sin(\alpha)). \quad (2)$$

The arising the near-field interference pattern can be measured via near-field scattering in the far field [1]. The consideration presented above is valid for stationary laser radiation. Below we will consider the formation of an interference pattern in the TSG interferometer using an ultrashort laser pulse. In this case, the interference pattern in the TSG interferometer can arise only if the following condition is satisfied: the travel time of the plasmonic wave from the groove to the slit, $t(x)=d(x)/v_{sp}$, where v_{sp} is the group velocity of the plasmonic wave, should be shorter than the duration $\delta\tau$ of the laser pulse. Hence, the duration of the pulse can be found from the expression $\delta\tau = (d_0 + X_m\sin(\alpha))/v_{sp}$ where X_m is the maximum value of the coordinate on the axis of the slit at which the interference pattern is still observed. The main idea of this work is to measure the duration of femtosecond laser radiation by measuring the interference pattern that it forms in a TSG interferometer.

Our calculations showed that the interferogram (cross-section of a 2D interference pattern) of continuous laser radiation has a pattern characterized by periodically modulated intensity with decreasing amplitude of the modulation. The amplitude decreases along the x-axis. The decrease in the modulation depth is related to the exponential decay constant of the plasmonic wave in gold, given by $\exp(-2k''_{sp}d(x))$, and it is determined by the characteristic time $t_{sp} = 1/(2k''_{sp}v_{sp}) \approx 160$ fs. For the laser radiation with a pulse duration shorter than time t_{sp} this behavior becomes much more noticeable. For a very short laser pulse with a duration of only $\delta\tau_0 = 6$ fs, the interferogram exhibits only three oscillation periods. In this limiting case, the effect of the decay of the plasmonic wave on the structure of the interferogram can be neglected, i.e., the decrease in the modulation amplitude is characterized only by the excitation dynamics of the plasmonic wave in the TSG interferometer, i.e. $\sim\exp(-d(x)/(\delta\tau_0 v_{sp}))$. Therefore, the shape of the interferogram strongly depends on the pulse duration of the laser radiation.

We can also solve the inverse problem, i.e., we can determine the laser pulse duration from the measured interferogram. In order to achieve this, it is necessary to find the decay of the interferogram intensity and to retrieve the pulse duration from the decay constant (exponential factor of the interferogram envelope). We would like to emphasize that the measurements of laser pulse duration with use of the plasmon interferometer is related to measuring of the first order autocorrelation function of the pulse. Pulse duration in this case can be obtained only in the assumption of a Furrier-limited laser pulse, i.e. in the same way as in measurements with the use of Michelson interferometer or other basically equivalent split and delay arrangements [13,14].

3. Experimental setup and samples

In this work, the TSG interferometers were prepared in a single-crystal gold nanofilm with a thickness of 200 nm via the ion-beam lithography method using a tightly focused beam of Ga^+ ions. The gold film (from PHASIS, Geneva, Switzerland) was obtained via

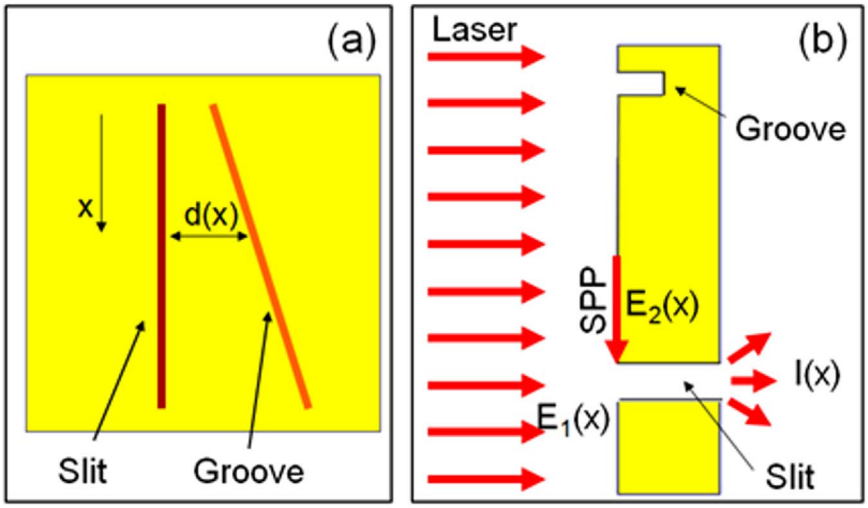

Fig. 1. Schematic of the tilted slit-groove interferometer: (a) top view, (b) excitation scheme.

epitaxial growth on the surface of a 100-μm-thick substrate. The electron microscopy of the gold film showed single-crystal flakes on the surface with a typical size of 3–5 μm. The quality of the nanofilm at the atomic level is of fundamental importance in nanoplasmonics [15,16]. First, this ensures control of the TSG interferometer geometry with a high spatial accuracy (in polycrystalline films, the accuracy of the geometry is defined by the crystal size of gold crystals, and is in the range of 20–50 nm). Second, unlike with polycrystalline films, there are no losses related to the scattering of electrons at interfaces between the nanocrystals, the characteristic size of which is 20–50 nm. The latter circumstance yields a large propagation length of surface plasmon polaritons in the TSG interferometer.

On a single-crystal gold nanofilm, different TSG interferometers with different parameters were made, i.e., with different lengths of the slit and groove, groove depth, and slit width. The spacing between the TSG interferometers was 200 μm, which allowed the exciting laser radiation impinge a single TSG interferometer.

The experimental setup was as follows. Laser radiation from a Ti-sapphire laser with pulse duration of 6 fs (two periods of the light wave) illuminates a sample on which TSG interferometers have been prepared. The sample was placed in the specimen plane of an inverted microscope. The duration of the laser pulse was varied using bandpass filters, which were placed in the path of the laser beam. As a source of continuous laser radiation, we used a Ti-sapphire laser with a central wavelength of 800 nm. The TSG interferometry was investigated using an inverted Nikon Eclipse microscope. The light transmitted through the slit of the TSG interferometer was collected with a mirror objective (40, NA = 0.5), and was analyzed with a CCD camera or spectrometer equipped with a CCD camera (Hamamatsu c9100-13).

The spectrum of a two-cycle laser pulse extends from 650 nm to 1 μm. Such a large spectrum width requires dispersion control of the medium over the whole propagation path from the laser to the sample. Experimentally, the group velocity dispersion was controlled with an accuracy of up to 0.1 fs^2 using an optical system that involved the following: (a) two dielectric mirrors, multiple reflection from which provided the necessary negative group velocity dispersion (equal to -250 fs^2) and (b) a system of optical wedges that provided positive group velocity dispersion in the range of 0–250 fs^2.

The group velocity dispersion and laser pulse duration were measured using a commercially available ultrashort pulse measurement technique - the SPIDER system. During the measurements, the nonlinear crystal of the SPIDER system and the sample with plasmonic nanostructures were placed at the same distance from the laser with an accuracy of ± 1 cm (the nanostructures were in the object plane of the microscope). This allowed us to guarantee with an accuracy of up to 0.2 fs that the laser pulse duration on the sample was the same as that measured by the SPIDER system.

Fig. 2a shows the electron microscope image of a TSG interferometer with the following parameters, slit: 100 nm × 200 μm, groove: 200 nm × 200 μm with a groove depth of 100 nm; and angle $\alpha = 15°$. Fig. 2b shows the interference pattern obtained from such an interferometer. The pattern was obtained through the transmission image of the TSG interferometer illuminated by a collimated continuous wave polarized laser beam. The maximal contrast of the interference pattern was obtained when the TSG interferometer was oriented such that the polarization vector of the laser radiation was nearly parallel to the interferometer slit. This orientation of the polarization corresponds to the minimal transmission of light by the nanoslit. The spatial period of the interference pattern was 3 μm, which corresponds to the value determined by the analytical expression (2).

The interference pattern obtained with the use of the TSG interferometer can be used to measure optical constants of metal nanofilms. Indeed, spatial period of the interference pattern can be used to measure real part of the dielectric permittivity of the film, while the decay constant can be used to measure imaginary part of

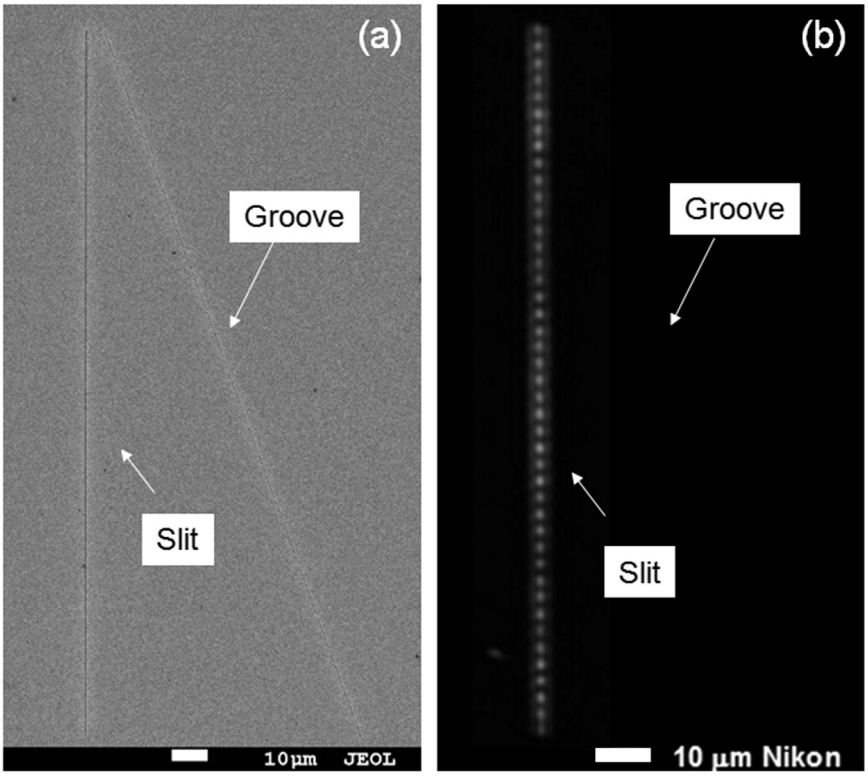

Fig. 2. (a) Electron microscope image of a TSG interferometer, (b) optical image (obtained in transmission mode) of a TSG interferometer that was illuminated by a collimated and polarized continuous wave laser beam at a wavelength $\lambda = 800$ nm.

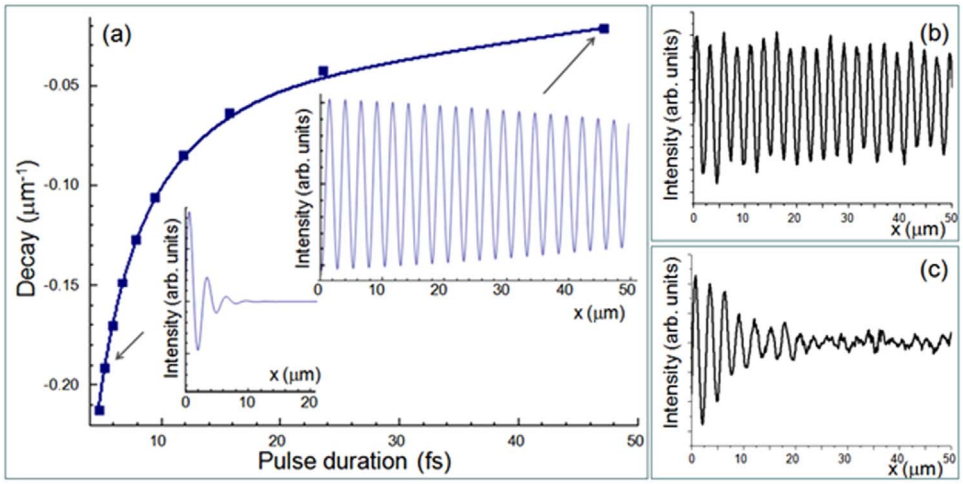

Fig. 3. Measurement of the duration of a femtosecond pulse using a plasmonic interferometer: (a) calculated dependence of the interferogram decay constant of the plasmonic interferometer on the laser pulse duration (insets show the interferograms determined for 6 fs and 50 fs pulses); measured interferograms for laser radiation pulses with durations of (b) 50 fs and (c) 6 fs.

the dielectric permittivity of the film. We have measured optical constants of the monocrystalline Au film with the use of the technique. In the visible spectral range the measured constants was in a good agreement with the data taken from Palik's paper [17]. We have made the extensive measurements with our plasmon interferometer including the characterization of many types of metal nanofilms: monocrystalline, polycrystalline, the films with different methods of their formations and etc. We have find out that TSG plasmon interferometry is possible with the use of a monocrystalline-type of the nanofilms only. The main reasons are as follows: (1) the large length of SPP propagation can be achieved only on single-crystal nanofilms and (2) in the case of polycrystalline nanofilms there are additional parasitic SPP sources due to scattering of SPP waves on metal nanofilm morphology imperfections.

4. Results

Fig. 3a shows the calculated dependences of the interferogram decay constant of the TSG plasmonic interferometer on the laser pulse duration that arises upon irradiation of the interferometer by a pulsed Ti-sapphire laser radiation at different pulse durations. The insets present the interferograms for pulses with durations of 6 and 50 fs. In the calculations, the TSG interferometer on the gold film with the following parameters was considered: angle $\alpha = 15°$, width $d_0 = 250$ nm. In our calculations, expression (1) was used to determine the interferogram for each spectral component of the laser pulse. After that, the summation of the interferograms obtained for each spectral component was performed. The dielectric constants of Au were taken from [17]. As it can be observed from Fig. 3, the decay constant of the interferogram of the TSG interferometer strongly depends on the duration of the laser pulse. We would like to emphasize that in the interval 5–20 fs, the slope of the decay constant curve is significantly large, much larger than the expected noise, and corresponds to the range of maximal sensitivity of the interferometer in the measurement of the laser pulse duration. The expected noise in the measured interferogram is the result of two main contributions: (1) imperfections of metal nanofilm that lead to a change of spp waves phase as well as to form a second-type sources of spp waves, (2) spatial inhomogeneity of laser light radiation.

We performed the TSG interferometry measurements with a laser light of different pulse durations, controlled using the SPIDER system. Our measurements have shown that for laser pulses with a duration in the range of 6–50 fs, the interferograms obtained show good agreement with the calculations presented in Fig. 3a. In Fig. 3b and c, we show the corresponding sections of the TSG interferograms obtained using two laser pulses with durations of 50 and 6 fs. The decay constant determined from the interferogram presented in Fig. 3c is equal to $k_D = -0.19\ \mu m^{-1}$, which corresponds to the value of the decay constant for the pulse with a duration of 6 fs. It should be noted that the number of interference maxima in the measured interferogram (Fig. 3c) is twice that in the calculated interferogram. We relate such a discrepancy to the occurrence of final length of the interferometer, which was not taken into account in our calculations. However, the presence of these maxima has no effect on the decay constant of the interferogram. We noted that there are considerable fluctuations in the intensity modulation amplitude in the interferograms. We assume that this behavior is caused by the occurrence of inhomogeneities on the surface of the gold film, which are secondary sources of plasmonic waves, that contribute to the interference of plasmonic waves from the slit.

5. Conclusion

In summary, we realized a plasmonic TSG interferometer for measuring the duration of femtosecond pulses in the range between 6 and 50 fs. The TSG interferometer is prepared using ion- or electron-beam lithography; therefore, it can be created directly on the sample with plasmonic structures to be examined. This makes it possible to use it in experiments as a local probe of the femtosecond laser pulse duration. The measured values of the laser pulse duration were confirmed using a commercially available ultrashort pulse measurement technique – the SPIDER system.

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