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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: $\mathrm{d}(x) = d_0 + x\sin(\alpha)$; 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: $E_2(x) \sim \exp\left(-k''_{sp}\mathrm{d}(x)\right)$ and to a similar dependence of its phase: $\Phi(x) = k'_{sp}\mathrm{d}(x) + \phi_0$. 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:

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

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:

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

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, $t(x)=\mathrm{d}(x)/v_{sp}$, 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 $\delta\tau = \left(d_0 + X_m\sin(\alpha)\right)/v_{sp}$ 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 $\exp\left(-2k''_{sp}\mathrm{d}(x)\right)$, and it is determined by the characteristic time $t_{sp} = 1/\left(2k''_{sp}v_{sp}\right) \approx 160$ 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. $\sim\exp\left(-\mathrm{d}(x)/\left(\delta\tau_0 v_{sp}\right)\right)$. 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

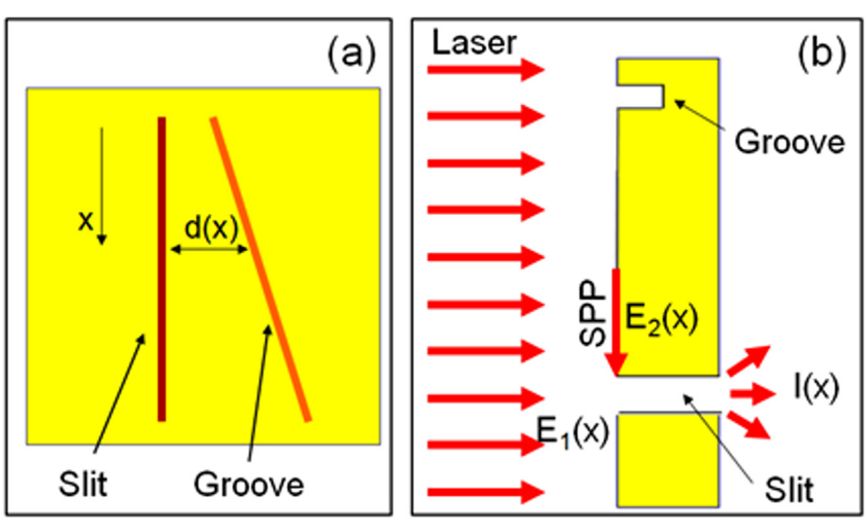

Fig. 1. Schematic of the tilted slit-groove interferometer: (a) top view, (b) excitation scheme.