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Femtosecond Surface Plasmon Interferometry with Gold Nanostructures

Vasily V. Temnov and Keith Nelson
Department of Chemistry, Massachusetts Institute of Technology, Cambridge, Massachusetts 02139, USA
Phone: (617) 452-2594, Fax: (617) 253-6295, e-mail: temnov@mit.edu

Gaspar Armelles and Alfonso Cebollada
Instituto de Microelectrónica de Madrid (CSIC), 28760-Tres Cantos, Madrid, Spain

Tim Thomay, Alfred Leitenstorfer, and Rudolf Bratschitsch
Department of Physics and Center for Applied Photonics, University of Konstanz, D-78457 Konstanz, Germany

Abstract: We measure the ultrafast electron dynamics in gold via ultrafast surface plasmon interferometry. A new plasmonic micronterferometer with tilted slit-groove pair is used to unambiguously determine changes of real and imaginary parts of the dielectric function.

OCIS Codes: 240.6680, 320.2250

Pioneering ultrafast time-resolved optical experiments with surface plasmons have demonstrated acoustic phonon generation in impulsively heated metal films [1] and sub-picosecond dynamics of the electron gas in gold [2]. However, owing to the phase-matched surface plasmon excitation mechanism in the applied Kretschmann configuration it is difficult to clearly distinguish between changes of the real and imaginary parts of the complex dielectric function of the metal. An analysis requires high-precision time- and angular-resolved reflectivity measurements [1] with angular dependencies strongly varying as a function of optical wavelength [2, 3].

Here, we present an experimental method of ultrafast surface plasmon interferometry, which combines the advantages of conventional time-resolved interferometry [4] with a new geometry of a plasmonic slit-groove microinterferometer. It is now possible to directly and unambiguously distinguish between the small transient changes of real and imaginary parts of the dielectric function of the metal excited by femtosecond laser pulses.

Our plasmonic microinterferometer consists of a 50 µm long, 200 nm wide, and 100 nm deep groove and a 50µm long 100nm wide slit milled with a focused ion beam into a 200 nm thick gold film on glass (Fig. 1). A 200 fs pump pulse at 400 nm (rep. rate 250 kHz, average power 160 µW) focused between the slit and the groove induces an ultrafast perturbation or the electron distribution in the gold layer. Time-delayed probe pulses at 800 nm (200 fs, average power 50 mW) homogeneously illuminate the entire microinterferometer under an angle of incidence of 10 degrees. Whereas nearly 98% of incident probe light is reflected from the gold surface, a small fraction is converted into surface plasmons due to the non-conservation of momentum at the sharp edges of the nanostructures. Ultrashort surface plasmon pulses generated at the groove propagate towards the slit and experience pump-induced changes of the dielectric function on their way. At the slit they are reconverted into free-space electromagnetic radiation and interfere with probe light directly transmitted through the slit. Due to 15° tilt angle between the slit and the groove a pronounced periodic interference pattern is observed along the slit:

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

The pump-induced transient perturbations of electronic distribution in gold are so small that no noticeable change of *I(x)* is observed as a function of pump-probe delay time (see upper panel in Fig. 1c). Therefore, we have developed a setup for scanning lock-in based detection of modulated probe transmission (modulating the pump beam at 1.5 kHz using a mechanical chopper) that preserves a high spatial resolution along the slit and allows for recording the pump-probe interferogram I_{pp}:

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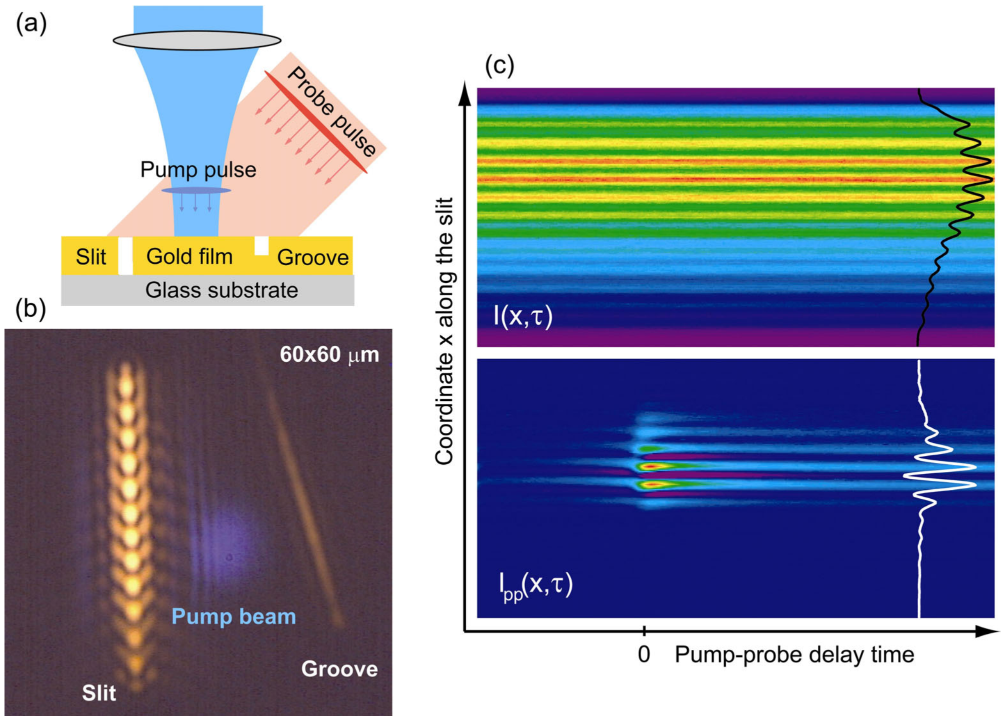

Fig. 1: (a) Pump-probe excitation geometry, (b) transmission image of the new tilted slit-groove microinterfeometer, homogeneously illuminated by ultrashort probe pulses and excited by a femtosecond pump pulse, focused between groove and slit. (c) Plasmonic interferogram I(x,τ) and pump-probe interferogram I_{pp} (x,τ). The pump-probe delay time varies between -3 and +5 ps.

$$I_{pp}(x) \approx -E_1 E_2 \frac{k_0 L_{\text{int}}}{|\varepsilon|^2}(d\varepsilon'' \cos\Phi + d\varepsilon' \sin\Phi),$$

which is shown in the lower panel of Fig. 1c. Here, L_{int} is an effective interaction length of the order of pump spot diameter. Simultaneous Fourier-based analysis [5] of the plasmonic interferogram I(x) and the pump-probe interferogram I_{pp} (x,τ) allows for the extraction of pump-induced changes of the real and imaginary parts of dielectric function with high spatial and temporal resolution [6]. We find an exponential relaxation of hot electrons due to electron-phonon interaction within 900 fs after excitation resulting in an increase of surface temperature of about 20 K in the center of laser-excited area.

We believe that our interferometric experiment for ultrashort surface plasmon pulses represents a powerful technique to tackle many problems in modern non-linear, quantum and magneto-plasmonics.

[1] M. van Exter and A. Lagendijk, "Ultrashort surface-plasmon and phonon dynamics," Phys. Rev. Lett. 60, **49** (1988)
[2] R. H. M. Groeneveld, R. Sprik, and A. Lagendijk, "Ultrafast relaxation of electrons probed by surface plasmons at a thin silver film," Phys. Rev. Lett. **64**, 784 (1990)
[3] J. Wang, J. Wu, and C. Guo, "Resolving dynamics of acoustic phonons by surface plasmons," Opt. Lett. **32**, 719 (2007)
[4] V. V. Temnov, K. Sokolowski-Tinten, P. Zhou, A. El-Khamhawy, and D. von der Linde, "Multiphoton Ionization in Dielectrics: Comparison of Circular and Linear Polarization," Phys. Rev. Lett. **97**, 237 403 (2006).
[5] M. Takeda, H. Ina, S. Kobayashi, "Fourier-transform method of fringe-pattern analysis for computer-based topography and interferometry", J. Opt. Soc. Am. **72**, 156 (1982)
[6] V.V. Temnov, K. Nelson, G. Armelles, A. Cebollada, T. Thomay, A. Leitenstorfer, R. Bratschitsch, to be published