Full Spatio-Temporal Control on Nanoscale

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1. Introduction

This NIRT Project is devoted to a major goal of creating a **nanoplasmonic portal**, that is an approach and device to coherently connect the far zone of optical fields with the near zone. This portal will allow one to transfer energy and coherence of optical fields from macroscopic to nanoscopic scale and exert **full spatio-temporal control of optical fields on nanoscale.** The main, original principle of the project is the adiabatic transformation of the surface plasmon polaritons (SPPs) in graded nanostructures from micro- to nanoscale [1-3]. The main such nanostructure will metal wedge whose transverse size slowly changes from microns to nanometers, which adiabatically compresses the SPP fields as the SPP approach the tip of the wedge (see Fig. 1 for illustration).

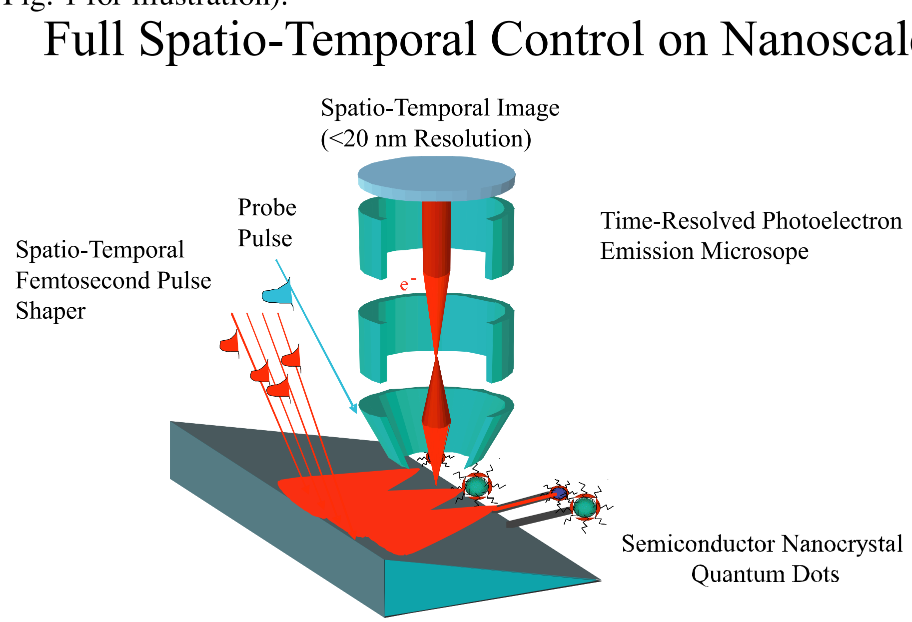


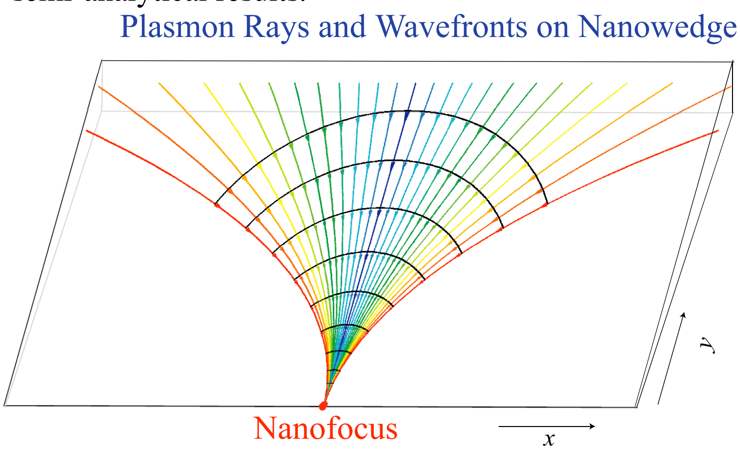
Fig.1. Nanoplasmonic portal: Adiabatic energy concentration on nanoscale

2. Developments and Results

The Project activities at Georgia State University (GSU) have been devoted to the theoretical consideration of the underlying adiabatic transformation of ultrafast optical fields from the far zone to the near zone. An important component of these activities has been support and theoretical guidance of the Project's experimental programs.

We have theoretically simulated the processes in nanoplasmonic portal where SPPs run from a wide end of a wedge toward its sharp tip. We have explored different wavelengths and dielectric environment of the wedge. The optimum concentration of optical energy occurs for a wedge in vacuum at a (vacuum) wavelength of 400 nm, in which case the energy concentration occurs at a scale of ~15 nm. This simulation promoted the development of a spatio-temporal modulator at the 400 nm wavelength.

We have theoretically investigated the required spatio-temporal modulation of the excitation pulses required for the functioning of the nanoplasmonic portal. While the full, numerical solution of the corresponding polaritonic problem is extremely difficult, we have employed the Wentzel-Kramers-Brillouin (WKB) approximation that allowed us to obtain robust and useful semi-analytical results.



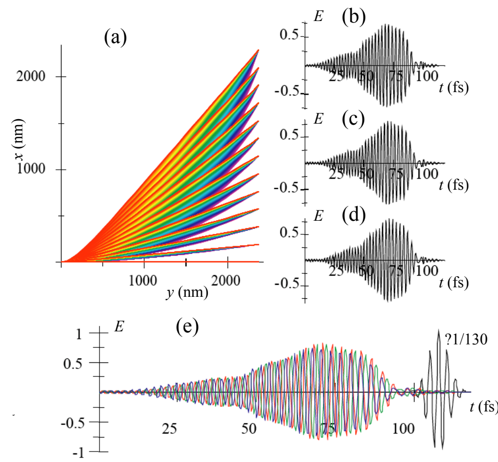


Fig.2. Plasmon polariton rays on the wedge portal converging toward the nanofocus and the corresponding wave fronts.

Fig.3. (a) Ultrashort excitation pulses converging toward the nanofocus. Trajectories are shown in the plane of the wedge. The colors indicate the corresponding frequency. (b)-(d) three temporally-modulated pulses that are applied to the two extreme and one intermediate points at the wedge, as shown. (e) The same pulses overlapping to show the phase relations (showed by colored lines) and the resulting ultrashort pulse at the nanofocus (black line).

We have undertaken a related theoretical research directed toward temporal coherent control on the nanoscale [3]. By changing the temporal delay between the two excitation pulses, it is possible to control the hot spot of the electron emission within 4 nm. Such spatial resolution is not achievable at the present time by any other means existing or proposed.

The following articles have been published or accepted for publication that enjoyed major funding from this project: [3-7].

The key element in the project is the transformation of spatiotemporally shaped optical waveforms with features of ordinary dimensions (microns or slightly less) into specified plasmonic waveforms with far smaller features (tens of nanometers) through adiabatic progression along a wedge-like structure with gradually diminishing metal thickness – our “nanoplasmonic portal” that connects conventional micro-optics with nano-plasmonics. The approach depends on experimental capabilities developed in the Nelson group for spatiotemporal femtosecond pulse shaping, in which a single light beam with a single femtosecond pulse is transformed into a shaped field with phase and amplitude profiles specified as functions of time and one spatial dimension. [8-10]. Spatiotemporal pulse shaping has been demonstrated and used extensively for 800-nm light.

Based on theoretical developments, we have decided to focus our initial efforts on implementation of the nanoplasmonic portal with incident light at 400 nm wavelength instead of 800 nm as planned originally. However, the spatiotemporal pulse shaping technology had only been demonstrated at 800 nm. Its extension to 400 nm required far more than simply changing the optics for the incident light, because the key element of the 800-nm pulse shaper is a 2D liquid crystal spatial light modulator (LC SLM). The liquid crystals absorb at 400 nm, so an entirely new apparatus was needed.

Fortunately a MEMS-based 2D SLM has recently been demonstrated [11] for temporal-only femtosecond pulse shaping, in which a shaped field is specified as a function of time but not position. The MEMS element consists essentially of a 2D array of small mirrors on pistons. These can be used for reflection of wavelengths across a wide spectral range extending well into the UV. We have put together a 400 nm optical setup for long-term use at this wavelength. This will enable generation of shaped waveforms that are optimized for transformation through the nanoplasmonic portal.

Metal wedges were fabricated by positioning the region of the substrate where the wedge was to be grown behind a small block of glass. The metal deposition rate onto the substrate is proportional to the area of evaporating metal in the boat "seen" by the substrate, leading to the formation of a tapered metal film in the shadow of the block (see Fig. 4). The width of the wedge can be controlled by adjusting the height of the block, while the slope is a function of the amount of metal deposited. Depending on the metal and the growth conditions, root mean square values for the surface roughness below 3 nm can be achieved. Wedges with predefined shapes can be fabricated by covering the substrate with a resist layer that was patterned by optical or electron beam lithography.

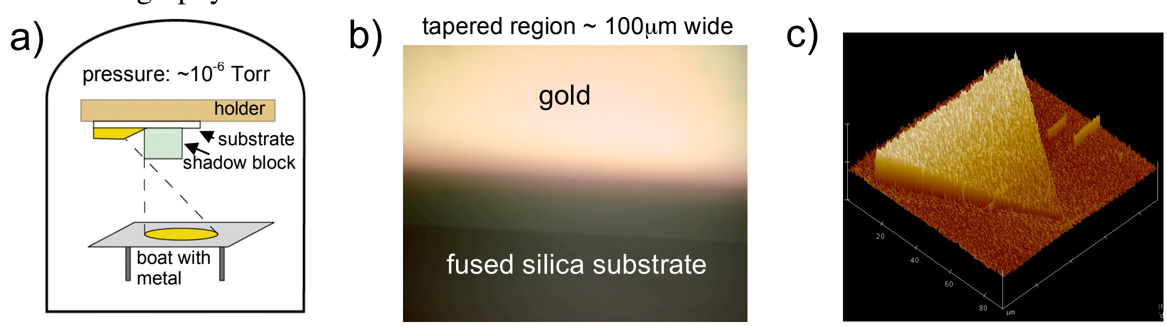


Fig 1: a) Schematic of the shadow evaporation setup used for fabricating metal wedges, base pressure ~ 10^{-6} to 10^{-7} Torr. b) Image of a gold wedge on a fused silica substrate taken with an optical microscope. c) Atomic force microscope image of a triangular gold wedge. The metal thickness increases form ~5nm to ~100nm over a distance of about 80um.

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