Femtosecond Energy Concentration in Nanosystems Coherently Controlled by Excitation Phase Modulation

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ABSTRACT

We predict and quantitatively evaluate the unique possibility of concentrating the energy of an ultrafast excitation of a nanosystem in a small part of the whole system by means of coherent control (phase modulation of the exciting ultrashort pulse). Such concentration is due to dynamic properties of surface plasmons and leads to local fields enhanced by orders of magnitude. This effect exists for both "engineered" and random nanosystems. We also discuss possible applications in nano-photonics, nano-lithography, and nano-computing

Keywords: nanoscale, ultrafast, optics, energy concentration, coherent control

1 INTRODUCTION

In this paper, we present theoretical prediction of a new effect that manifests itself as femtosecond concentration of the optical excitation energy in nano-structured systems [1]. This concentration occurs spatially in a small part of the nanosystem. The specific localization site is controlled by phase modulation of the exciting ultrafast laser pulse.

There recently has been a tremendous interest in both ultrafast (femtosecond and attosecond) laser-induced kinetics and in nanoscale properties of matter. Particular attention are attracted by phenomena that are simultaneously nanoscale and ultrafast, see, e.g., Refs. 2-5. Fundamentally, nanosize eliminates effects of electromagnetic retardation and thus facilitates coherent ultrafast kinetics. On the applied side, superdense (on the nanoscale) design of optoelectronic elements is justified if their operating times are ultrashort to allow for ultrafast computing and ultra-wideband transmission of information.

One of the key problems of ultrafast nanoscale physics is ultrafast excitation of a nanosystem where the transferred energy localizes at a given site or propagates along a given path. Because electromagnetic wavelength is on the much larger microscale, it is impossible to employ light-wave focusing for that purpose. Our idea to solve this problem is to use phase modulation of an exciting femtosecond pulse as a functional degree of freedom to coherently control spatial distribution of the excitation energy. This possibility exists due to the fact that polar excitations (surface plasmons) in inhomogeneous nanosystems tend to localize with their frequency correlated with position [6]. In that case, changing phase of the excitation will cause the exciting field to take energy from excitations in some parts of the system and to add energy to surface plasmon oscillations of other parts as desired.

The coherent control (phase modulation of the pulse) has been recently used to concentrate energy of an ultrashort nonlinear-generated pulse in a given high harmonic [7]. Our effect is based on the same general idea of interference between different components of the exciting radiation interacting with a resonant system governed by the phase modulation, but it is a linear effect in contrast to Ref. 7.

2 THEORY

Quantitative theory of this effect is based on the Green's function method. The local field potential $\varphi(r,t)$ is expressed in terms of the exciting field potential $\varphi_0(r,t)$ via a retarded Green's function $G^r(\mathbf{r}',\mathbf{r};t)$ as

$$\varphi(\mathbf{r},t)=\varphi_0(\mathbf{r},t)-\int\varphi_0(\mathbf{r}',t')\frac{\partial^2}{\partial\mathbf{r}'^2}G^r(\mathbf{r}',\mathbf{r};t-t')\,d^3r\,dt. \quad (1)$$

This Green's function is expressed as the spectral expansion over eigenmodes $\varphi_n(\mathbf{r})$ and eigenvalues s_n of the system in the coordinate-frequency domain,

$$G^r(\mathbf{r}',\mathbf{r};\omega)=\sum_n\varphi_n^*(\mathbf{r}')\varphi_n(\mathbf{r})/\big(s(\omega)-s_n\big), \quad (2)$$

where $s(\omega)$ is the spectral parameter, a property of the system's material but not of its geometry [8]. The eigenmodes $\varphi_n(\mathbf{r})$ and eigenvalues s_n, which depend only on the system's geometry but not on its material properties, have been computed by numerically by solving the quasi-electrostatic boundary problem [9]. The temporal

dependence in Eq. (1) has been computed by fast Fourier transform.

3 NUMERICAL RESULTS

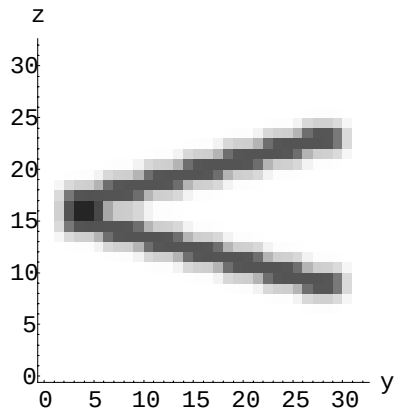

Fig.1: Geometry (density) of nanosystem (V-shape). The spatial coordinates are on the nanoscale (realistically, one unit may be 2-5 nm).

The nanosystem studied is an "engineered" V-shape shown in Fig.1, positioned in the yz plane. The computations have been made for silver as a material. The exciting pulse is z polarized and has the Gaussian envelope with the unit maximum value,

$$\varphi_0(\mathbf{r},t) = -z\exp\left[i\omega_0\left(1+\alpha\frac{t-T/2}{T}\right)(t-T/2)-\frac{3}{2}\left(\frac{t-T/2}{T}\right)^2\right]+\text{c.c.}, \quad (2)$$

where ω_0 is the average frequency of the pulse, α is the phase modulation parameter, T is the characteristic pulse length.

The spatial distribution of local electric fields at the nanosystem at different moments of time is shown in Fig. 2. The exciting pulse and temporal dynamics of electric fields at certain characteristic sites of the nanosystem is displayed in Fig. 3. Note that the position of the pulse on the temporal scale is arbitrary, but consistent between the figures.

As one can see from Fig. 2(a), at the beginning of the pulse ($t = 51.8$ fs) the excitation field and energy are concentrated at the wider part of the nanostructure (the "opening" of the V-shape). When the pulse progresses ($t = 60.2$ fs), as seen in Fig. 2(b), the region of excitation (polarization) moves toward the vortex of the V-shape. This snapshot corresponds to the maximum of the pulse in Fig. 3(b). After the exciting pulse ends ($t = 104$ fs), as shown in Fig. 2(c), *almost all excitation is concentrated at a narrow, nanometer-scale site*, close to the tip of the V-shape. Separate computations (data not displayed) have shown that in the absence of the phase modulation (for $\alpha = 0$), it is impossible to achieve such a localization of energy for any frequency ω_0. The present phenomenon reminds somewhat the "Ninth Wave" effect,[4] with an important difference: in Ref. 4 the energy concentration occurs due to giant fluctuations caused by the randomness of the system, while here this nanoscale localization is due to the effect of coherent control (the chosen phase modulation of the exciting pulse).

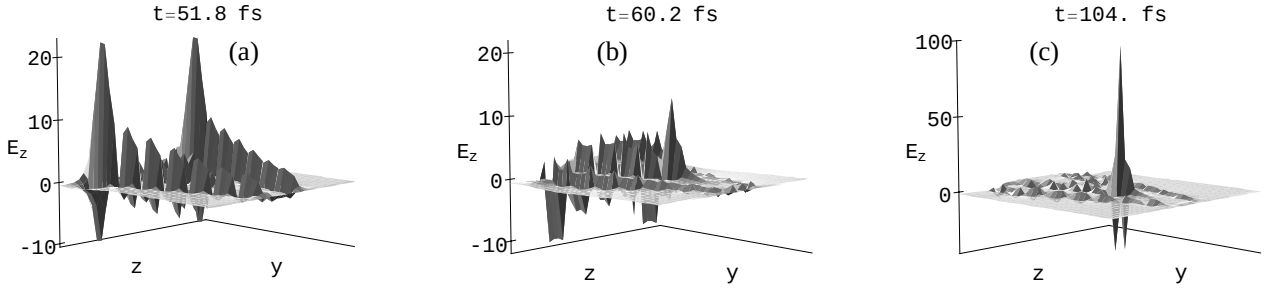

Fig.2. Spatial distributions of local electric fields (in the units of the exciting pulse amplitude) at the nanostructure for moments of time indicated. The duration of the pulse is 25 fs, the phase modulation parameter $\alpha = 0.3$, and the average frequency $\omega_0 = 0.95$ eV.

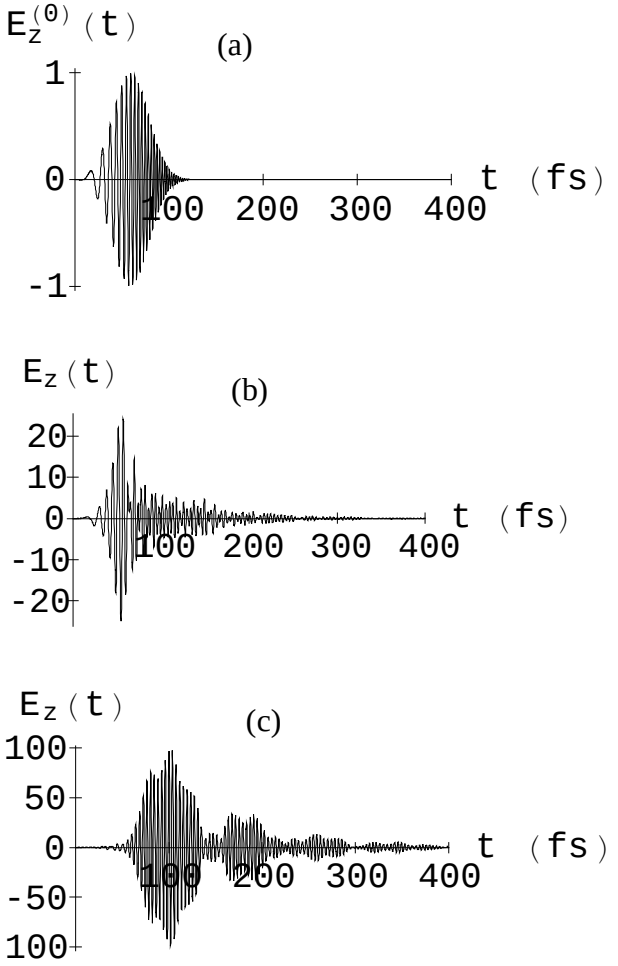

Fig. 3a: Temporal dependence of the exciting pulse. Fig. 3b: Dynamics of the local field at the point of (local) maximum in Fig. 2(a). Fig. 3c: Dynamics of the local field at the point of the global maximum in Fig. 2(c).

The effects of the phase modulation can be traced using temporal behavior of the local fields. As Fig. 3(b) demonstrates, after the maximum excitation at the initial excitation site (the V-shape widest area) us reached, the energy is actually "pump out" by the exciting pulse during just a few oscillations due to the established phase relation between the oscillation and the pulse. In contrast to that, much higher frequency oscillations established in Fig. 3(c) is in favorable phase relation with the pulse until it ends. Therefore this oscillation persists for much longer and is so high (four orders of magnitude enhancement in energy with respect to the exciting pulse). Note that polarization relaxation times corresponding to the dynamics in Fig. 3(c) are $\cong 10$ fs, which agrees on the order of magnitude with experiments of Ref. 3. The multiple revivals of coherence are due to the long time (≈ 1.7 ps) of the energy dissipation in noble metal nanoparticles known experimentally from Ref. 5.

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