Photoelectron Emission from Resonant Nanoantennas Driven by Femtosecond Mid-infrared Pulses

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Abstract: Strong-field photoelectron emission from gold nanorod antennas induced by mid-infrared pulses is studied using time-of-flight spectroscopy. The emission and acceleration of photoelectrons are maximized at the half-wave antenna resonance, evidencing substantial near-field enhancements.
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1. Introduction

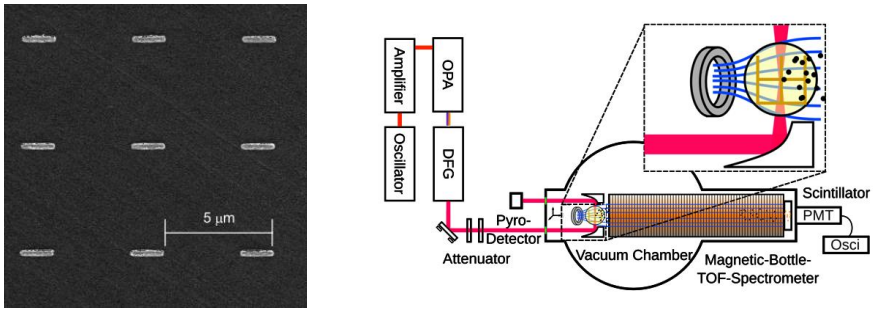
The enhancement of electromagnetic fields with sub-wavelength confinement achieved with metal nanostructures offers extensive potential for the study of strong-field light-matter interactions, including field-driven photoelectron emission [1-4] and other nonlinear optical processes. Compared to the near-infrared spectral range, driving strong-field phenomena at longer wavelengths has the attraction of larger ponderomotive potential $U_p=e^2E^2/4m\omega^2$ and less material damage from linear and multi-photon electronic excitation.

Here, we investigate photoelectron emission from resonant gold nanorod antennas driven by mid-infrared femtosecond pulses. The electron yield displays a pronounced enhancement at the half-wave antenna resonance. The intensity dependence of the photoemission yield evidences mid-infrared field enhancements, which cause substantial ponderomotive acceleration. The observed intensity enhancement factor on resonance is estimated to lie above a value of 2000.

2. Experimental Setup

Metal nanorods of few-micron length exhibit their half-wave antenna resonance in the mid-infrared spectral range [5]. Here, gold nanorods of different lengths (from 1.0 to 2.5 μm) are fabricated on ZnS substrates by electron beam lithography and a lift-off process. Individual nanorods are arranged in 2D arrays as shown in Fig.1(a). Both the lateral and vertical distances between neighboring units are set to 5 μm, in order to avoid possible near-field couplings.

Widely tunable few-cycle mid-infrared laser pulses (λ = 1~10 μm) are generated by difference frequency mixing of signal and idler waves from an optical parametric amplifier, pumped by amplified Ti:Sa laser pulses (rep. rate 1 kHz, central wavelength 800 nm, pulse duration 50 fs). Electron emission from gold nanorod arrays is induced by focusing the mid-infrared pulses using an off-axis parabolic mirror. Photoelectron yields and kinetic energy spectra as a function of wavelength and intensity are measured with a magnetic-bottle time-of-flight spectrometer.



(a) (b)

Fig. 1. (a) A scanning electron micrograph of a nanorod array and (b) the experimental setup for photoemission spectroscopy.

3. Photoelectron Emission from Resonant Nanorod Antennas

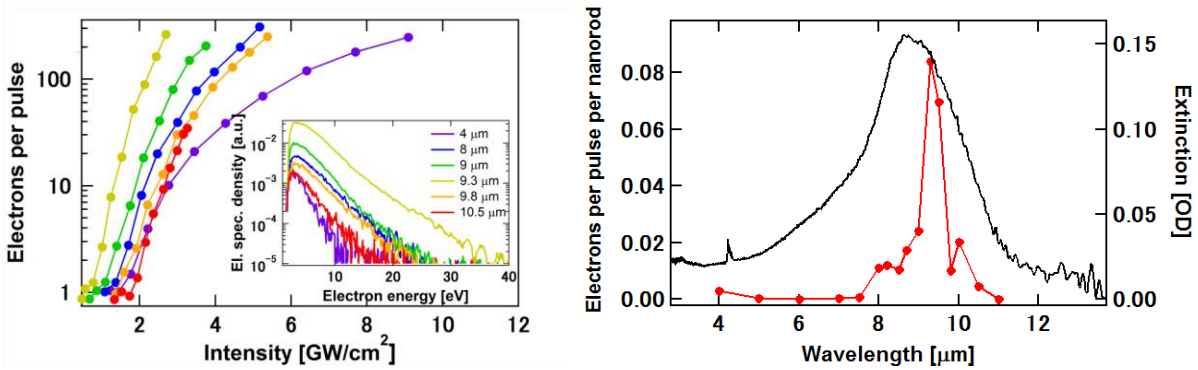
Metal nanorods display local field enhancements at their half-wave antenna resonance. The enhancement is prominent in the vicinity of the antenna edge, and decays rapidly with a characteristic length of sub-100-nm scale along the antenna axis. This promotes optical field emission and subsequent light-driven acceleration of electrons at substantially reduced photoemission thresholds.

Figure 2 (a) shows the intensity dependence of the electron yield for different wavelengths. At resonance, significant photoemission is observed at excitation intensities as low as 1 GW/cm^2. When electron tunneling from nanoantennas occurs as the potential barrier is distorted by the local field, the intensity enhancement factor η enters the Fowler-Nordheim tunnel equation in the following way [6]:

$$I = \frac{e^3 \eta E^2}{16\pi^2 \hbar \phi t^2(y)} \exp\left(-\frac{4\sqrt{2m_e}}{3\hbar}\frac{\phi^{\frac{3}{2}} v(y)}{e\sqrt{\eta}E}\right). \quad (1)$$

Here, I, E and ϕ are the current density, the electric-field amplitude and the metal work function, respectively, and $t(y)$ and $v(y)$ are slowly varying functions accounting for the Schottky effect. The intensity enhancement factor is estimated to be above 2000 for a resonant excitation at a wavelength of 9 μm. This value agrees with predictions from finite-difference time domain (FDTD) simulations. Such large enhancement is characteristic for nanoantennas resonant in the mid-infrared, because of high (geometric) aspect ratio of the structures and reduced non-radiative/radiative damping at lower frequencies [7]. The kinetic energies of photoelectrons measured for constant peak intensity (~2.4 GW/cm^2) are maximized at 9.3-μm excitation wavelength (Fig.2a, inset) and reach several tens of eV, demonstrating near-field-driven electron acceleration.

Figure 2 (b) compares an optical far-field extinction spectrum (solid line) of a nanorod (length 2 μm, ZnS substrate) with the photoemission yield measured with laser pulses of constant peak intensity (2.7 GW/cm^2) at each wavelength (circles). The electron yield displays a pronounced enhancement at the half-wave antenna resonance. For such antennas, the maximum near-field enhancements occur typically at lower frequencies than the peak of the far-field spectrum [8], which results in a weak red-shift of the photoemission peak, as shown in Fig.2(a).



(a) (b)

Fig.2. (a) Intensity dependence of electron yields for different excitation wavelengths (inset: kinetic energy spectra measured for constant peak intensity of ~2.4 GW/cm^2). (b) The far-field extinction spectrum for 2-μm-long nanorods (black) and the photoemission yield (red).

4. Conclusions

We have studied the emission and acceleration of photoelectrons from resonant gold nanoantennas driven by mid-infrared femtosecond pulses. The spectral dependences of the electron yield and kinetic energy spectra evidence substantial near-field enhancements at the antenna resonance, with the local intensity enhancement factors above 2000 obtained from wavelength-dependent Fowler-Nordheim fits.

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