Few-Cycle Laser Pulse induced Plasmon Assisted Thermionic Injection in Metal-Insulator-Metal Junctions

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Abstract: Gold nanoparticles on a metal-insulator-metal junction locally enhance the absorption of few-cycle laser pulses. The locally heated electron gas leads to thermionic emission exceeding multiphoton emission and allows detection of single nanoparticles.
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1. Introduction

In the interaction of intense laser pulses with nanostructures or optical antennas the electron emission is commonly discussed in the context of multiphoton processes [1] and strong field phenomena [2–4]. However, for sufficiently strong excitation, fast carrier thermalization, and small enough work function the local electronic system is heated to high temperatures leading to dominant thermionic emission. Qualitatively, thermionic emission is well described by the Richardson-Dushman equation. For a work function of 2-3 eV an electron gas temperature of several thousand Kelvin is required to obtain sufficiently high emission current densities. Surprisingly this excitation regime has not yet been demonstrated for few-cycle laser pulse excitation. This is explained by the fact that for homogeneous excitation of a surface such high electron gas temperatures also lead to substantial lattice heating and corresponding thermal surface degradation, i.e. unstable emission conditions. However, local resonant excitation and the related field enhancement also significantly increase the locally absorbed energy density. The spatially inhomogeneous excitation reduces thermal degradation and thus opens the possibility to detect thermionic emission.

Here we demonstrate that few-cycle laser pulse excitation of a localized surface plasmon in a single metal nanoparticle leads to a strong spatially localized electron gas heating in the supporting layer, i.e. the top electrode of a metal-insulator-metal (MIM) junction, and induces thermionic injection currents in the MIM junction. The fast heat diffusion out of the highly localized excitation spot (∅ ≈ 100 nm) within the time between subsequent laser pulses (≈ 10 ns) reduces the local thermal load and the junction remains stable under these illumination conditions.

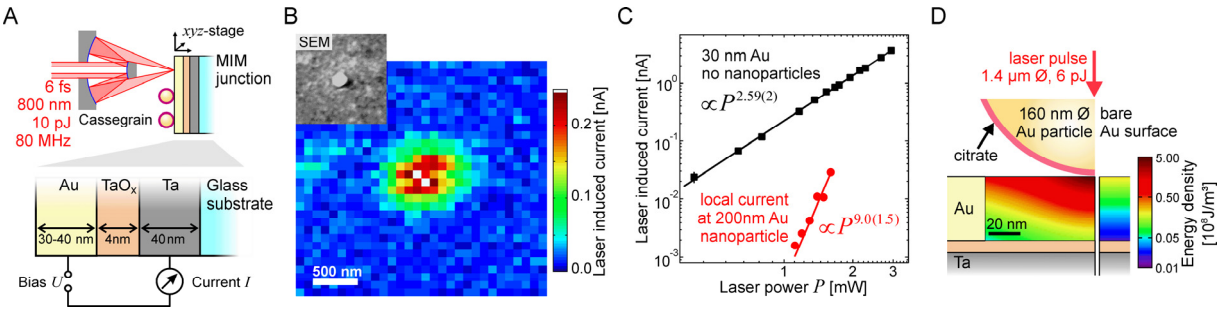

Fig. 1. Few-cycle laser pulse induced internal thermionic injection currents at Au nanoparticles supported on a Au-TaO_x-Ta metal-insulator-metal (MIM) junction. A) Scheme of the experimental setup. B) Spatial map of the local laser induced injection current in the MIM junction as the laser spot is scanned across the surface. The inset in the upper left corner shows a SEM image of the Au nanoparticle that is positioned in the centre of the scan area. The lateral scale of the SEM image is the same as in the current map. C) Power dependence of the laser induced current for a MIM junction with 30 nm thick Au top electrode (black squares) and a junction with 40 nm top electrode and excitation on top of a Au nanoparticle with 200 nm diameter (red circles). The solid lines represent power law fits and the exponents are indicated. D) Contour plot of the locally deposited energy density in the Au top electrode for excitation of a Au nanoparticle positioned on top of the layer (left part) and for excitation of the bare junction (right part) as derived from FDTD calculations.

2. Experimental setup

The experimental setup is shown schematically in Fig. 1A. The recently demonstrated scanning internal photoemission microscopy method [5] is used to investigate the local laser induced injection current in a metal-insulator-metal (Au-TaO_x-Ta) junction. Few-cycle laser pulses are focused onto the surface using a reflective

microscope objective. The sample is mounted on a *xyz*-nanopositioning stage and the laser induced currents are measured using a lock-in amplifier. Citrate stabilized Au nanoparticles of about 160 nm diameter were grown in aqueous solution and then dispersed on the surface by deposition of a few µl of solution on the junction and subsequent evaporation of the solvent. Combined scanning electron microscopy (SEM) and scanning microscopy of the reflected light is used to characterize the distribution of nanoparticles on the surface. Comparison of SEM images and reflected light maps of the same sample area allows identifying individual nanoparticles (inset in Fig. 1B) and perform laser induced injection current scans for an individual nanoparticle (Fig. 1B). Repeated scans for various incident laser power yields the intensity dependence of the locally induced currents (Fig. 1C) that are fitted by a power law to determine the nonlinear exponent.

3. Results and Discussion

For a bare MIM junction with a 30 nm thick Au top electrode a homogeneous injection current signal is measured across the surface. The intensity dependence (black symbols in Fig. 1C) yields a nonlinear exponent of 2.6 indicating a mixture of two-photon and three-photon processes that are responsible for the observed laser induced injection current as it is expected for the given MIM junction having an internal barrier height of 1.8 eV [5]. Increasing the top electrode thickness to 40 nm reduces this multiphoton induced homogenous injection current below the detection limit (≈ 1 pA) and hence these junctions are perfectly suited to investigate localized excitations induced by Au nanoparticles dispersed on the surface (Fig. 1B). The size of the nanoparticles is chosen to generate a plasmonic resonance close to the excitation wavelength for particles deposited on a gold substrate. The citrate shell serves as a thin insulation layer and the nanoparticles have no metallic contact to the substrate as seen in charging effects during scanning electron microscopy. In the laser induced injection current map the nanoparticles light up as highly localized spots with peak currents in the order of 0.2 nA (example for an individual particle is shown in Fig. 1B). Hence, metal nanoparticles on MIM junctions are interesting candidates for ultra-small and highly efficient nonlinear photo detectors for future photonic applications.

Interestingly the local injection current varies highly nonlinear with the incident laser power. The nonlinear exponent of 9.0(1.5) (Fig. 1C) cannot be attributed to above threshold processes since for them also the lowest possible nonlinear order dominates the total emission current since the contribution from higher order processes decreases exponentially [4]. The strong field emission regime leads to a reduced nonlinearity compared to the multiphoton regime and thus can also not account for the highly nonlinear intensity dependence. In contrast, following the Richardson-Dushman equation thermionic emission generates highly nonlinear intensity dependencies with the nonlinearity varying strongly as the heating is increased. As derived from a FDTD calculation a gold nanoparticle deposited on a metal-insulator-metal junction enhances the excitation of the top metal layer by a factor of 50 compared to the bare junction (Fig. 1D) leading to locally deposited energy densities in the order of 10^8-10^9 J m^{-3}. Based on the electronic heat capacity of Au this corresponds to local electron gas temperature of about 3000 K. Assuming an electron cooling time constant of about 1 ps and an effective excitation spot radius of 50 nm yields a local thermionic emission current of about 0.2 nA and a local nonlinear slope very close to 9. Thus, the simple thermionic emission model quantitatively explains both the measured laser induced current magnitude and the observed nonlinearity. An electron relaxation cascade model, based on electron-electron and electron-phonon scattering rates, allows simulating the multiphoton and thermionic emission over an internal tunnel barrier. Variation of both scattering rates determines the ratio between sequential multiphoton photoemission and thermionic emission following the Richardson-Dushman equation. Choosing realistic values of the model calculations confirms the qualitative model based on the Richardson-Dushman equation alone.

4. References

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