

Ultrafast nanoplasmonic photoelectron dynamics between the multiphoton and strong-field regimes with tunable IR excitation

Balázs Bánhegyi^{1,2}, Gábor Ligeti¹, Zsolt G. Kiss¹, Zsuzsanna Pápa^{1,3}, Péter Rácz¹, and Péter Dombi^{1,3}

1. Wigner Research Centre for Physics, 1121 Budapest, Hungary

2. Budapest University of Technology and Economics, 1111 Budapest, Hungary

3. ELI-ALPS Research Institute, 6728 Szeged, Hungary

We examined transitions between multiphoton-induced photoemission and strong-field tunnelling of electrons at the surface of gold nanoparticles using infrared femtosecond pulses between 800 nm and 2.4 μm with our newest experiments. In addition to the observation of non-adiabatic tunnelling [1], we establish the theoretical background of plasmonic excitation in the infrared range, calculate spectra by solving the time-dependent Schrödinger equation (TDSE), and then verify the predictions with experimental results.

For our investigations, we used 70-fs pulses from an optical parametric amplifier (OPA) output between 800 nm and 2.4 μm , with 400 nm intervals. The photoemission target was a plasmonic gold nanorod array with the nanoparticle size of 570x120x50 nm³ on a fused silica substrate with indium-tin-oxide (ITO) conducting layer. To obtain the plasmonic properties of the nanorods, we performed finite-difference time-domain simulations. In these simulations, we also considered the slight roughness of the gold nanorods based on SEM images (Fig. 1 inset). The emitted electrons were detected using a time-of-flight (ToF) spectrometer.

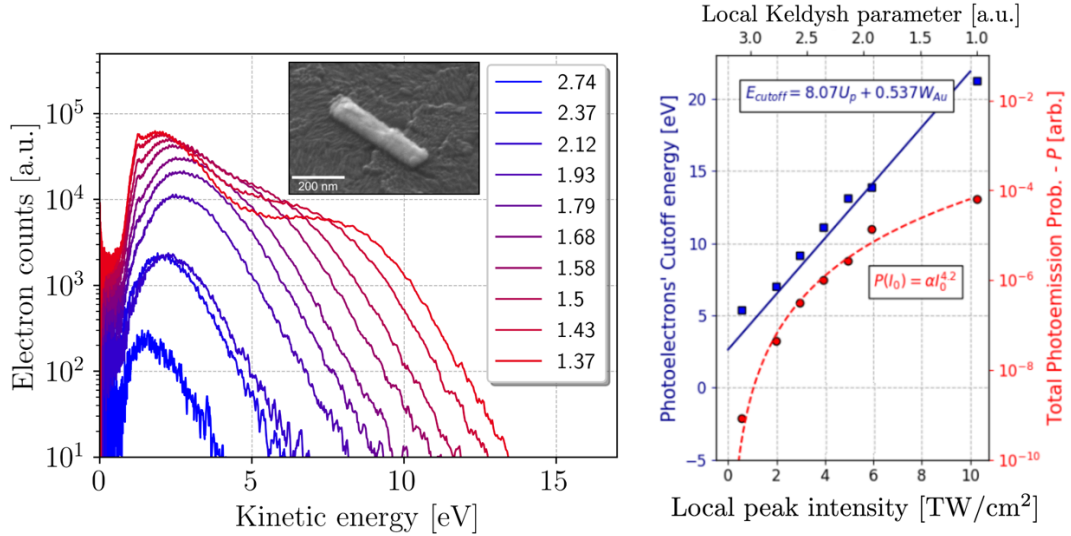


Fig. 1. (Left) Measured kinetic energy spectra of the emitted electrons at increasing local peak intensities using 1.6 μm femtosecond pulses. (Inset) SEM image of a single plasmonic nanorod in the nanostructured array used as photoemission target. (Right) Calculated cutoff energies and electron yields as a function of local peak intensities.

Looking at the measured data, the characteristic change in the shape of the emission spectra with increasing local intensities due to the multiphoton - strong-field transition is clearly visible also indicating a drastic change in the emitted electron dynamics. To verify the transition process, we extracted the cutoff electron energies for the spectra and plotted them as a function of the local peak intensity calculated from the incident intensities and the plasmonic field enhancement values. We defined the cutoffs as the kinetic energy below which 99.5 % of the electron yield is contained as a cutoff definition [2]. This method allows us to define cutoff energies for the triangular-shaped (multiphoton-like) spectra as well. The observed cutoff trend results in a $9.8(\pm 2)U_p$ scaling all cases, signifying the presence of the above-mentioned rescattering events even when spectra have a triangular shape, and no plateau is visible. This suggests that both multiphoton and tunnel emission is present in our experiments resulting in hybrid photoemission channels.

References

[1] Béla Lovász, Péter Sándor, Gellért-Zsolt Kiss, Balázs Bánhegyi, Péter Rácz, Zsuzsanna Pápa, Judit Budai, Christine Prietl, Joachim R. Krenn, and Péter Dombi, “Nonadiabatic Nano-optical Tunneling of Photoelectrons in Plasmonic Near-Fields,” *Nano Lett.* **22**, 2303 (2022)

[2] Timo Paschen, Ryan Roussel, Lennart Seiffert, Björn Kruse, Christian Heide, Philip Dienstbier, Joshua Mann, James Rosenzweig, Thomas Fennel, and Peter Hommelhoff, “Ultrafast strong-field electron emission and collective effects at a one-dimensional nanostructure,” *ACS Phot.* (2023)