Deviations from Integer Power Laws in fs-Laser Excited PEEM Due to Coexisting Electron Emission Mechanisms

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The observation of non-integer (i. e. fractional) power laws observed in fs-laser excited PEEM has been a puzzle [1,2] because it is obviously in conflict with the concept of multiphoton photoemission (nPPE) predicting $I_{electron} \sim I_{photon}^{n}$ (n being integer). In this contribution we discuss results for well-defined Ag nanocluster films deposited on Si (characterized by SEM, AFM and HiRes-TEM). Excitation by the fundamental (hν=1.45-1.65eV) and first harmonic (hν=2.9-3.3eV) of a Ti:sapphire laser excludes 1PPE due to the workfunction of the clusters being $\geq$3.8eV. The cluster films deposited as stepped wedges appear bright in the PEEM images, see Fig.1a. Corresponding cluster sizes as determined by HR-TEM (see inset) vary from 3-4nm in stripe 1 up to >50nm in stripe 4. Stripe 5 is a percolated film followed by bulk silver.

Electron kinetic energy spectra have been measured as function of photon energy and intensity (i.e. power of the fs-laser pulse) using an imaging high-pass energy filter or alternatively a time-of-flight technique as described in [3]. Upon variation of the photon energy of the first harmonic, the Fermi level onset in the spectra shifts by $2\Delta h\nu$ in accordance with 2PPE, see Fig. 1b. However, the power dependence is non-uniform over the spectra, see Fig. 1c. We discuss several alternative emission mechanisms that may become significant for certain conditions. The transient "hot" (non-Fermi-Dirac [4]) electron gas confined in small clusters may cause possible changes in the optical constants, absorption coefficients, matrix elements of the electron transition probability and parameters in the Richardson-Dushman equation. Plasmon-mediated nPPE is special because the localized surface plasmon enhances the brightness in PEEM due to the near-field [5] and "saves" photons for several fs leading to a high concentration of electromagnetic energy in nanometer-sized clusters. Finally, optical field emission may occur at very high laser power (more precisely, local electrical field). For excitation at hν=1.45-1.65eV the local intensity vs laser power curves show different regimes with different non-integer slopes. Fermi level onset shifts by 2-2.5$\Delta h\nu$ when increasing the laser frequency and appears significantly smeared out in electron energy spectra.

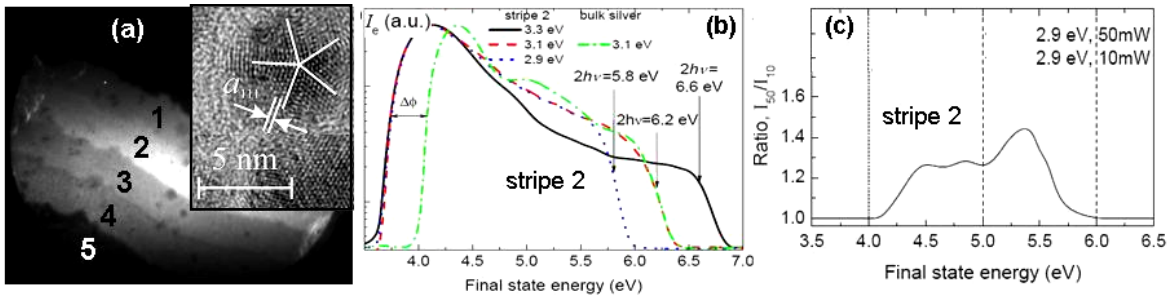

Fig.1: (a) *PEEM image of Ag cluster films with varying mass thickness (stripes 1 to 5 correspond to 1, 2, 5, 10 and 20nm, respectively) for fs-laser excitation at hν=3.1eV; the inset shows a HR-TEM image of two individual clusters.* ***(b)*** *Kinetic energy spectra of stripe 2 taken at different photon energies.* ***(c)*** *Ratio of energy spectra taken at hν=2.9eV with two different laser powers (normalized at the maximum at 4 eV).*

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