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# Proposal of Photoionization and Photofield Tunneling of Electrons as (Indirect) Evaporation Mechanisms during Laser-assisted Field Evaporation of Insulators

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Proceedings

# Proposal of Photoionization and Photofield Tunneling of Electrons as (Indirect) Evaporation Mechanisms during Laser-assisted Field Evaporation of Insulators

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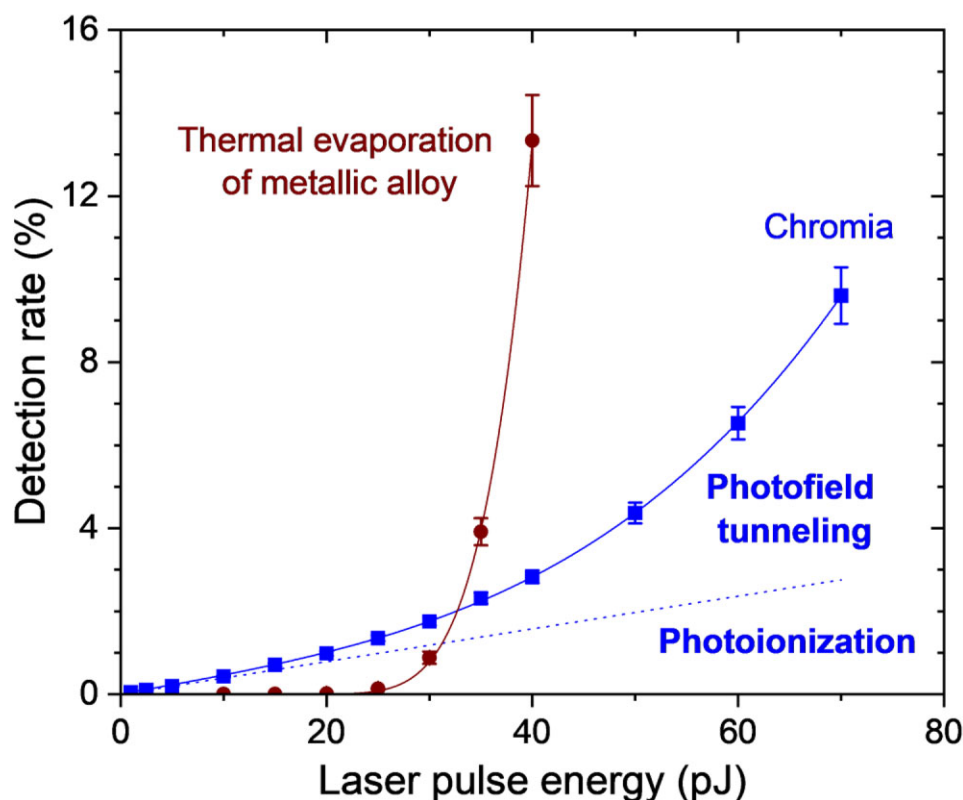
Atom probe tomography is a materials analysis technique that can attain highly localized chemical information with high sensitivity for all elements. It is based on field evaporation of ions in a high electrostatic field. To achieve this, a needle shaped specimen with an apex radius smaller than 100 nm is exposed to high voltages thereby creating electrostatic fields in the range of tens of V/nm. The sample is held at cryogenic temperatures during the experiment. For conducting materials an additional voltage pulse is used to ionize surface atoms towards a position-sensitive detector by time-of-flight mass spectrometry. The ion species is identified by time-of-flight mass spectrometry and an algorithm allows for the reconstruction of the evaporated volume. [1,2]

The use of short laser pulses instead of voltage pulses opened up the technique for non-conductive materials [3,4]. So far, the widely accepted mechanism has been thermal activation, i.e. heating the specimen and thereby inducing ion desorption. Not all observations can be explained by this. Several studies show a wide range of possible laser pulse energies (LPEs) in insulators with UV laser light [5–7]. Recently the authors reported on similar observations for chromia in the newest generation of APT instruments with deep UV laser light [8]. The low end of the LPE values seem too low for creating a significant thermal pulse. Similarly, APT measurements with extreme UV laser light could evaporate oxides but not metals and the heat input was considered to be too small for classical thermal activation [9,10].

We are presenting an experimental approach to investigate the evaporation mechanism. By keeping the standing voltage constant, the electrostatic field on the APT tip is held at a constant value. Varying the LPE results in a change in detection rate (DR). The influence of tip blunting can be mitigated by intermittently establishing the initial target DR before changing to a new LPE. More details can be found in [11]. It has been shown in literature that the peak temperature of the APT sample is approximately proportional to the LPE [12–14]. Applying this procedure on the metallic alloy Ni-20Cr shows the expected Arrhenius fit for thermal evaporation (see Fig. 1), validating the experimental approach. The same procedure applied to thermally grown chromia has revealed a rather different correlation between DR and LPE. At low LPEs the DR shows a linear regime, indicating photoionization. The non-linear part cannot reasonably be fitted to classical thermal evaporation.

In the absence of mobile charge carriers, the electrostatic field extends into the material and upon laser illumination, electron-hole pairs are created and the charge carriers become separated [15]. Electrons are conducted towards the stage and holes accumulate at the apex of the tip. Simulations indicate that a higher number of holes in the vicinity of a surface atom reduces the energy barrier for ion desorption [16]. We propose this as the critical step in field evaporation of insulators. Ion desorption is rate-limited by the generation and accumulation of new holes at the apex. New charge carrier holes are generated by photoionization as well as photofield tunneling of electrons. Experimental observations of the DR as a function of LPE during experiments with deep UV laser light on differently manufactured chromia as well as SrTiO<sub>3</sub> suggest a superposition of photoionization and photofield tunneling [11]. This is further supported by the correlation of the DR with field estimates during experiments with stepwise voltage changes. The efficiency of the hole generation seems to be significantly improved by photon energies above the work function. In that case, the photon energy is high enough to remove an electron from the material without additional energy input like the electrostatic field or temperature. Previously unexplained evaporation of MgO, Fe<sub>3</sub>O<sub>4</sub> and GaN in UV laser-assisted field evaporation shows the same dependence of the electrostatic field on LPE as chromia, suggesting that the same processes are in effect.

## Evaporation at constant voltage with deep UV laser pulses



**Fig. 1.** Detection rate as a function of laser pulse energy during experiments with intermittent constant standing voltage. The metallic alloy shows correlation with an Arrhenius fit, i.e. classical thermal evaporation. Experiments on chromia can be explained by the generation and interaction of charge carrier holes within the material.

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