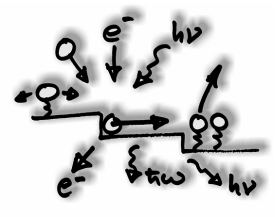
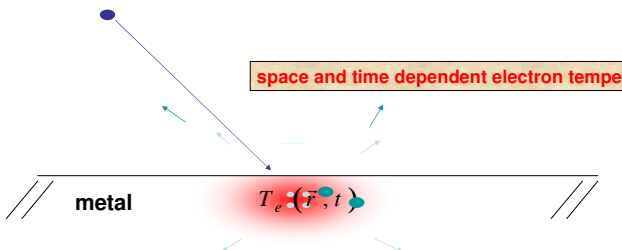
Electron promotion and electronic friction in ion-bombarded metals: a computer simulation model



A. Duvenbeck[1], O. Weingart[2], V. Buss[2] and A. Wucher[1]

[1]*Department of Physics, University of Duisburg-Essen, D-47048 Duisburg, Germany*
[2]*Department of Theoretical Chemistry, University of Duisburg-Essen, D-47048 Duisburg, Germany*

Motivation



- atomic motion in collision cascade — molecular dynamics simulation
- electronic excitation + excitation energy transport
- electron emission, charge state of sputtered particles

Goals

- Quantification of kinetic energy dissipation processes
- Calculation of space- and time-dependent excitation energy density within the collision cascade
- Determination of ionization probability for each sputtered particle

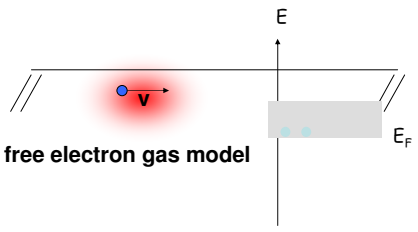
Model

Model = excitation mechanism + excitation energy transport

Lindhard friction

- electronic stopping:

$$\frac{dE_k}{dx} = -Kv \longrightarrow \frac{dE_k}{dt} = -Kv^2 = -AE_k,$$



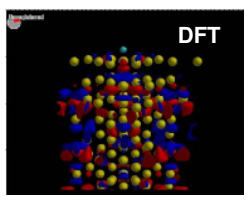


Fig. 1 : Trajectory snapshots of an H-atom impinging onto an Al(111) surface with kinetic energy of 10 eV [from M. Lindenblatt, University of Kiel]

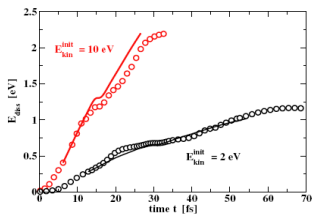


Fig. 2 : Energy E_{dis} dissipated into electron–hole pair excitations as a function of time for initial kinetic energies of the H-atom of 2 eV and 10 eV. The dissipated energy E_{dis} refers to a single hydrogen atom. The results from the TDDFT simulation (circles) are compared to the forecasts of the Lindhard theory.

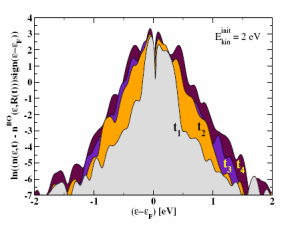
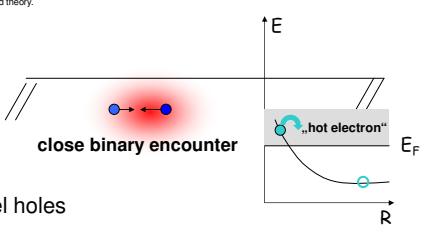


Fig. 3 : Snapshots of the time evolution of the excitation spectra for holes and electrons. The initial kinetic energy of each of the two H-atoms that impinge onto the Al(111) surface from either side of the slab amounts to 2 eV.

Electron promotion

- quasi-molecular orbital shift in close binary collisions
- promotion of 4d electrons to continuum states above Fermi
- production of hot electrons & localized deep level holes



Ag-Ag correlation diagram

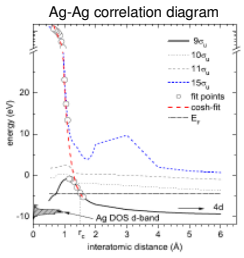


Fig. 4 : ab-initio calculation of mlecular orbital energies vs interatomic separation distance

resonant transitions

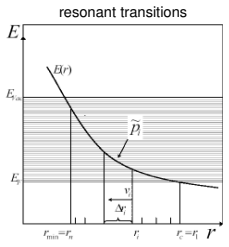


Fig. 5 : schematic drawing of a molecular orbital intersecting a conduction band

transition probability density

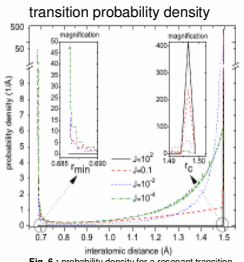


Fig. 6 : probability density for a resonant transition of a promoted electron from a MO into the conduction band

methodology

- construct diabatic orbital energy curve from ab-initio correlation diagram (DFT, Fig. 4)
- describe e^- - liberation via resonant transition from promoted 4d-level to continuum (Landau-Zener, Fig. 5)
- calculate probability distribution of excitation energies (stochastic, Fig. 6)
- consider case of weak coupling

transition most likely at distance of closest approach

Excitation energy transport

- nonlin. diffusion of excitation energy density
- excitation energy diffusivity D
- Neumann surface boundary condition

$$\frac{\partial E(\vec{r},t)}{\partial t} - \vec{\nabla}\cdot\left(D\left(T_l(\vec{r},t), T_e(\vec{r},t), \Lambda(\vec{r},t)\right)\vec{\nabla}E(\vec{r},t)\right) = A\sum_{i=1}^{N} E_k^i(\vec{r}_i,t)\cdot\delta(\vec{r}_i(t)-\vec{r}(t)) + \sum_{\kappa}\frac{E_{9\sigma_u}(r_{dca}^{(\kappa)}(t)) - E_F}{\Delta t}\cdot\delta(t-t_{dca}^{(\kappa)})\cdot\delta(\vec{r}-\vec{r}_{dca}^{(\kappa)})$$

$$D = D(T_e, T_l, \Lambda)$$

lattice temperature $T_l(\vec{r},t) = \frac{2E_k^{tot}(\vec{r},t)}{3N_{tot}k_B}$

electron temperature $T_{el}(\vec{r},t) = \sqrt{\frac{2}{C}E(\vec{r},t)}$

lattice order par. $\Lambda(\vec{r},t) = \frac{1}{3Z}\left|\sum_i\sum_{j=1}^{3}\cos\left(\frac{2\pi}{a_j}x_j^i(t)\right)\right|$

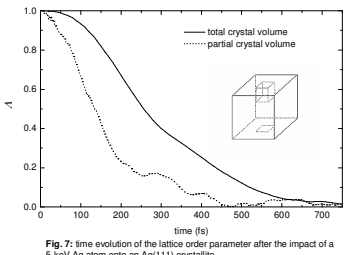
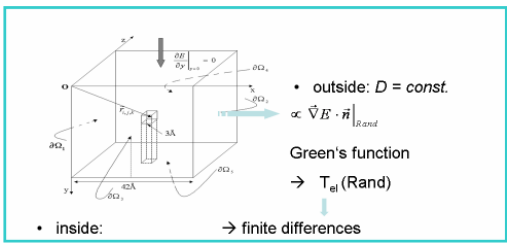


Fig. 7: time evolution of the lattice order parameter after the impact of a 5-keV Ag atom onto an Ag(111) crystallite

boundary conditions



Results

5-keV Ag Ag (111)

- simulated time: 750 fs
- MD/MC-CEM potential
- model crystallite 42 x 42 x 42 Å^3
- 4500 atoms

excitation energy densities at the surface

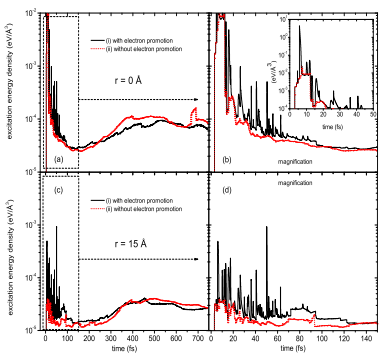


Fig. 9: Temporal snapshots of the exemplary collision cascade initiated by 5-keV Ag→Ag(111) bombardment. In addition to the illustration of atomic particles (blue: bulk atoms; green: surface atoms) the local electron temperature is visualized using the indicated colormap.

excitation energy density

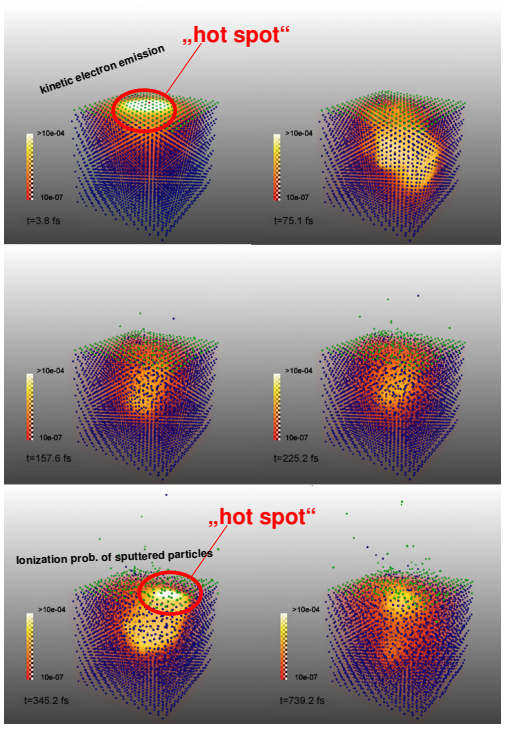


Fig. 8: Temporal snapshots of the exemplary collision cascade initiated by 5-keV Ag→Ag(111) bombardment. In addition to the illustration of atomic particles (blue: bulk atoms; green: surface atoms) the local electron temperature is visualized using the indicated colormap.

energy partition

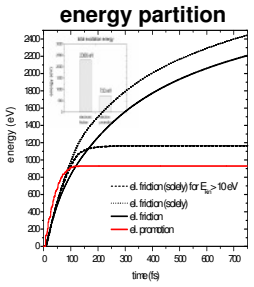


Fig. 10: **Volume-integrated excitation energy generated by electronic friction and electron promotion**

- **60% of the kinetic energy is dissipated into electronic degrees of freedom**
- **electron promotion dominating in the initial stage of the cascade (< 50 fs)**
- **two different kind of „hot spots“ of high excitation energy density**
- **local electron temperatures of about 1000 K at the surface during sputtering**
- **prediction of ionization and excitation probability for each sputtered atom possible using existing theory**

Recent Publications

[1] A. Duvenbeck, F. Sroubek, Z. Sroubek and A. Wucher, *Nucl. Instr. Meth. B* **225** (2004) 464-477
[2] S. Meyer, D. Diesing, and A. Wucher, *Phys. Rev. Lett.* **93** (2004) 137601-1 - 4
[3] A. Duvenbeck, Z. Sroubek and A. Wucher, *Nucl. Instr. Meth. B* **228,** (2005) 325-329
[4] A. Duvenbeck, M. Lindenblatt and A. Wucher**,** *Nucl. Instr. Meth. B* **228,** 170-175 (2005)
[5] A. Duvenbeck and A. Wucher, *Phys. Rev. B* **72** (2005) 165408-1
[6] M. Lindenblatt, E. Pehlke, A. Duvenbeck, B. Rethfeld and A. Wucher, *Nucl. Instr. and Meth. B* **246** (2005) 333
[7] A. Duvenbeck, O. Weingart, V. Buss and A. Wucher, *Nucl. Instr. And Meth. B* (submitted)
[8] A. Duvenbeck, O. Weingart, V. Buss and A. Wucher, *Phys. Rev. B* (submitted)

SFB 616

We acknowledge financial support from the Deutsche Forschungsgemeinschaft in the frame of the Sonderforschungsbereich 616 „Energy Dissipation of Surfaces“