Theoretical studies of ultrafast processes at metal surfaces .

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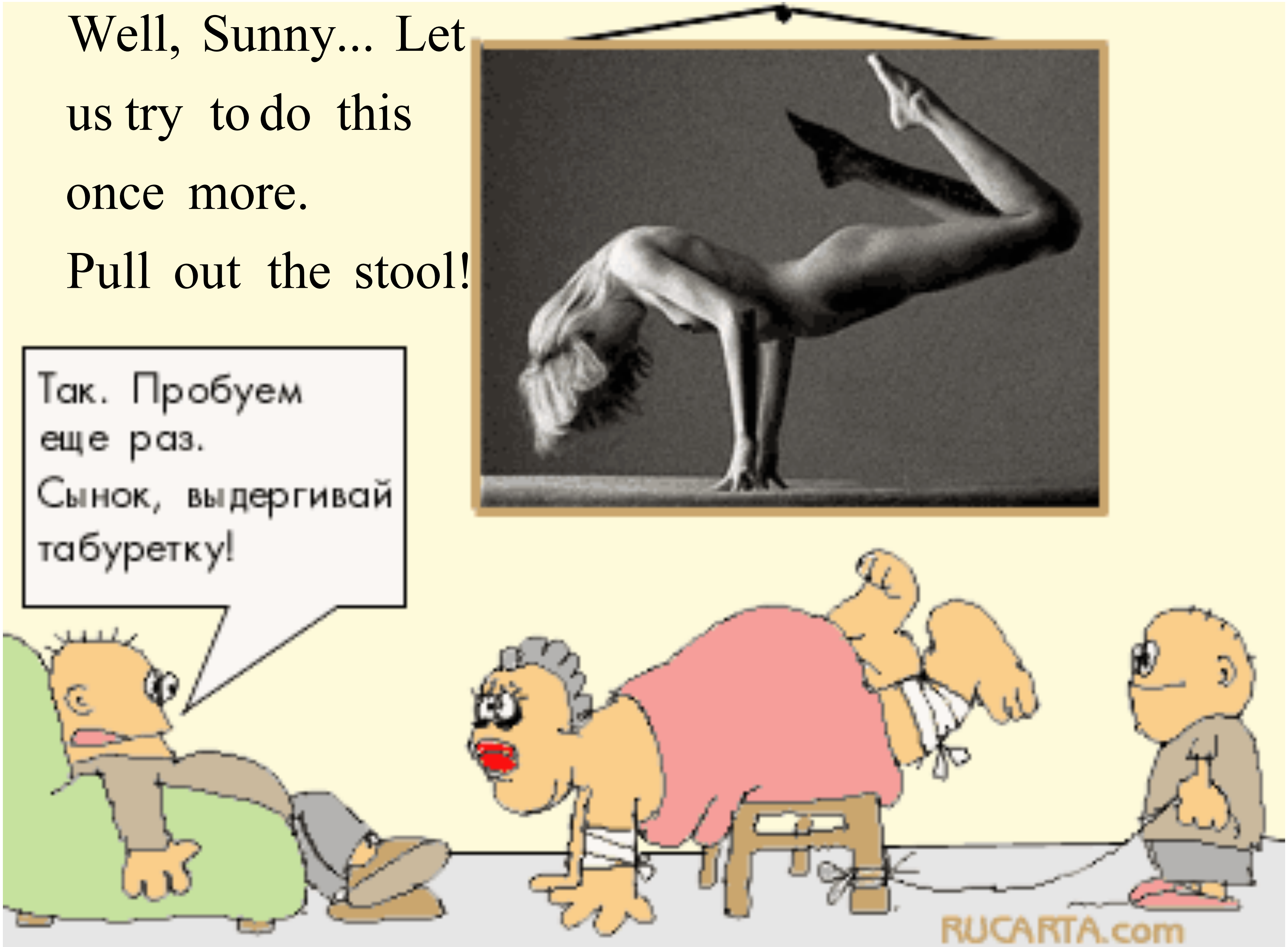
Well, Sunny... Let us try to do this once more.
Pull out the stool!



The principle features

1. The IR laser field in the metal is screened and its strength decreases promptly into the bulk.

2. The ejected electrons suffer inelastic collisions with electrons of the metal and this determines a depth from which the electrons can reach the surface without collisions and thus carry a direct information on the processes in the bulk.

3. The ejected by XUV pulse electrons move in the field by the lattice. Taking into account #1, does the band structure and the group velocity of the electron wave packet in the final state are physically meaningful?

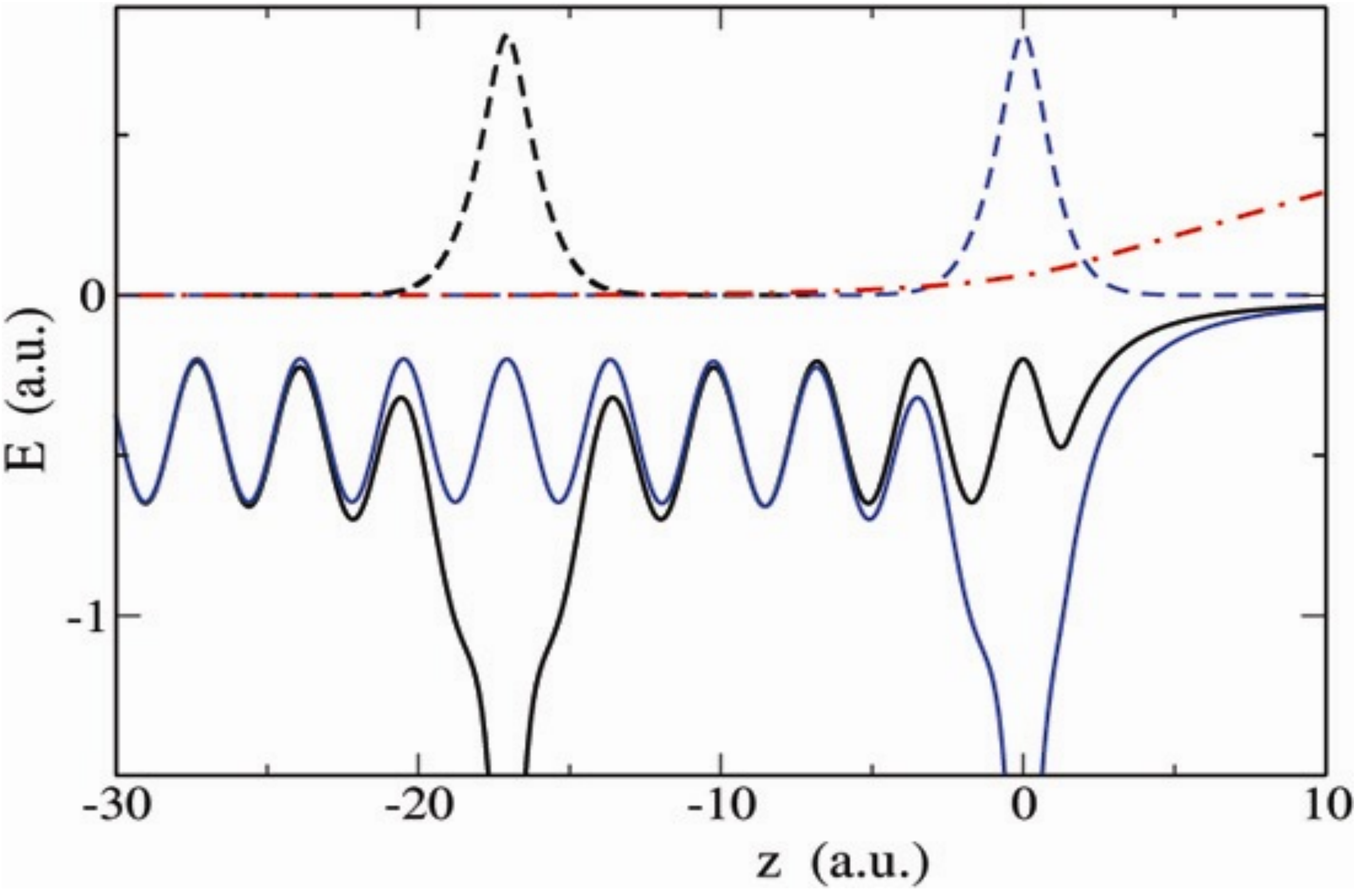
3. A localized electron after its ejection leaves in the bulk a positively charged hole which is then screened by the itinerant electrons. Could this screening be observed?

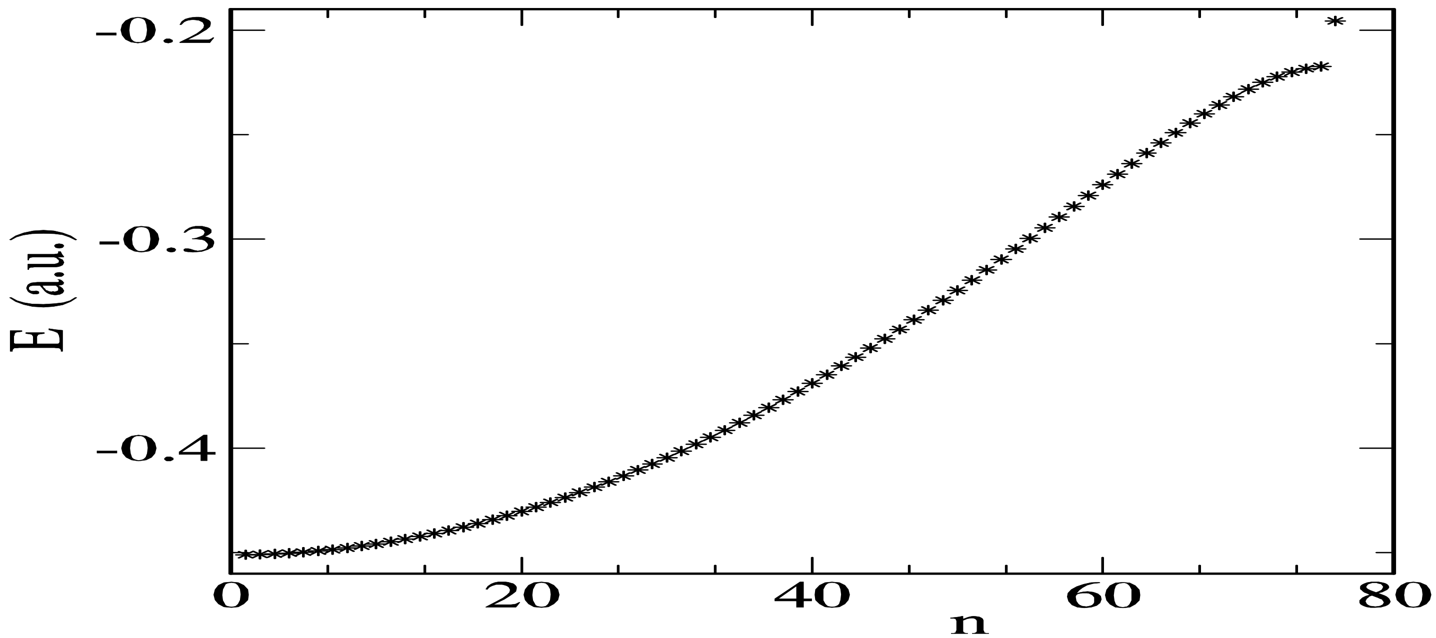
   And… what can be said about an delocalized electron???

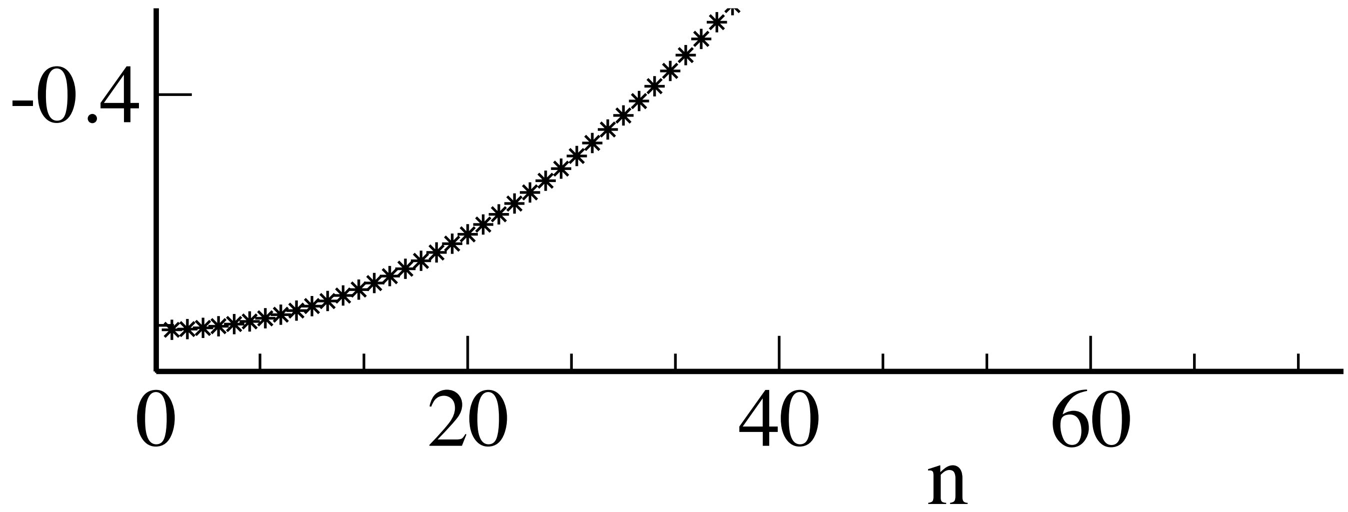
A one-dimensional model of streaking experiment with solids

A.K.Kazansky and P.M.Echenique.
Phys.Rev.Lett. 102, 177401, 2009

E = -1.565 a.u. E = -1.577 a.u.




-0.2
-0.3
-0.4
E (a.u.)
0
20
40
60
80
n


-0.4
0
20
40
60
n

Formalities

$$i\frac{\partial}{\partial t}\Psi(z,t) = \left(-\frac{1}{2}\frac{\partial^2}{\partial z^2} + U_s(z) - E + U_h(z - R_{at}) - i\gamma(z)\right)\Psi(z,t) +$$

$$E_L(t)f(z)\cos(\omega_L t)\Psi(z,t) + \frac{1}{2}E_{XUV}(t)z\Phi_0(z); \qquad \Psi(z,-\infty) = 0.$$

$$U_h(z) = -\frac{\exp(-|z - z_{at}|/\xi)}{\sqrt{(z - z_{at})^2 + (0.4)^2}}; \qquad \gamma(z) = \begin{cases} 0 & z > z_{im} \\ \Gamma/2 & z < z_{im} \end{cases}, \qquad \Gamma = \frac{\sqrt{2E}}{\lambda_f};$$

$$f(z) = \begin{cases} (z - z_{im}) + \xi & z > z_{im} \\ \xi \exp((z - z_{im})/\xi) & z < z_{im} \end{cases};$$ P.J.Feibelman, P.R.L, 30, 975, (1973)

$$U_s(z) = Cu(111). \qquad z_{im} = 2.73\,a.u., \qquad \xi = 4\,a.u., \qquad \lambda_f = 10\,a.u.$$

$$s_n(\varepsilon) = 2\pi\sqrt{2\varepsilon}\left|\int_{-\infty}^{\infty} dz \exp(-ikz)\Psi_n(z, t = +\infty)\right|^2;$$

$$k = \sqrt{2\varepsilon}.$$

$$\text{COM}(t_{delay}) = \int d\varepsilon \sum_{n=0}^{16} s_n(\varepsilon)\,\varepsilon \Big/ \int d\varepsilon \sum_{n=0}^{16} s_n(\varepsilon)$$

The cases considered:

1. The initial state is localized, the final energy E = 2 a.u.
2. The initial state is localized, the final energy E = 3 a.u.
3. The initial states are delocalized, the final energy E = 3 a.u. for the central state of the band.

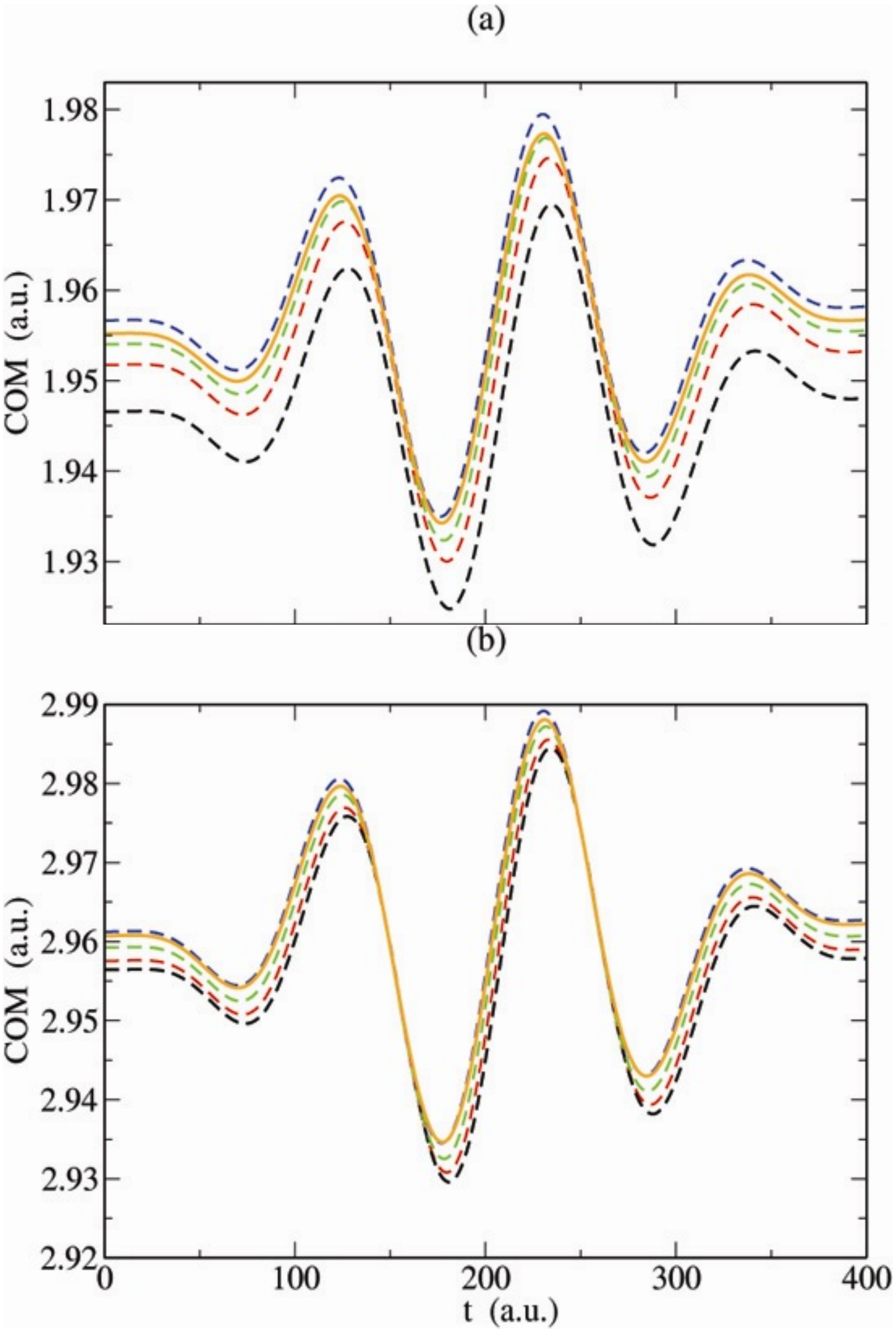
The frequencies of the XUV pulse for the cases 1. and 3. are very close.

Overview of the results

0. The necessary condition: $\lambda_f > \xi.$

- The spectra are almost Gaussian.
- The magnitude of the yield decreases exponentially: $I_n = I_0 \exp(-na/\lambda_f)$

3. The COM of spectra from localised states are almost uniformly shifted.



$$t' = t + \tau(z_{at}, E);$$

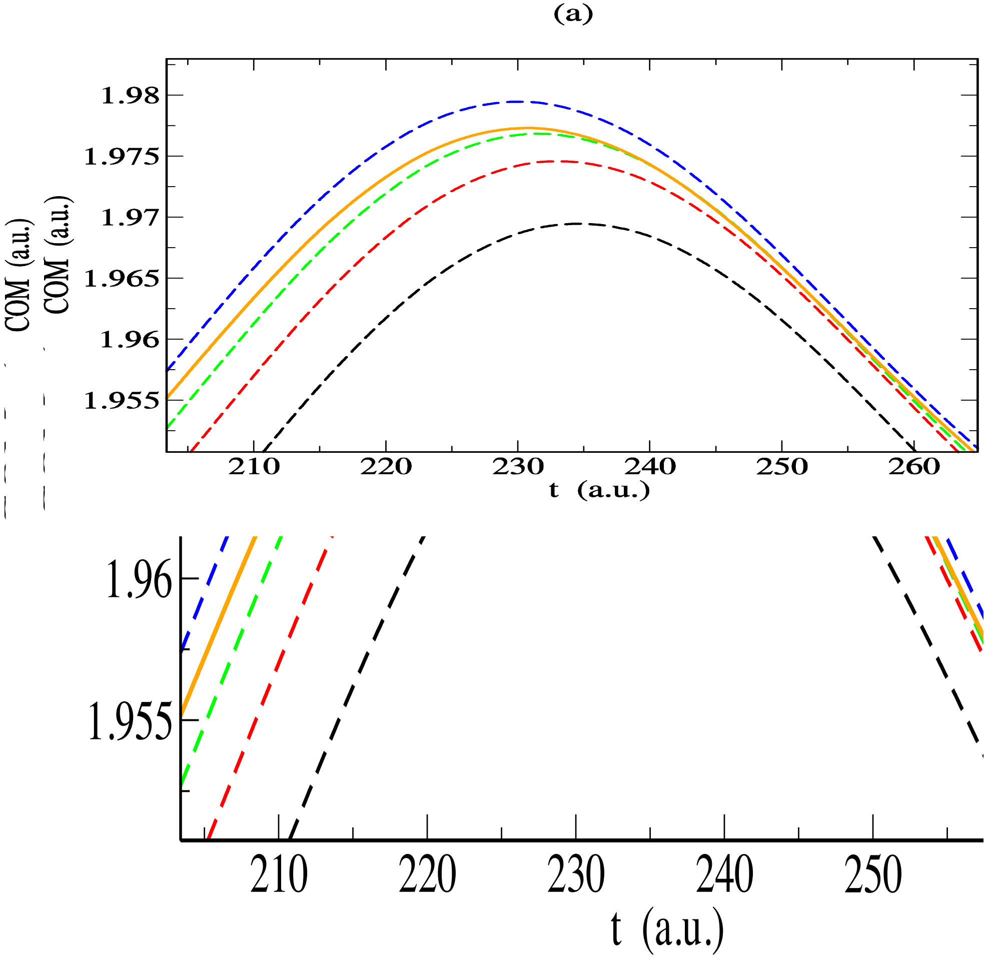
$$v(E) \equiv z_{at} / \tau(z_{at}, E);$$

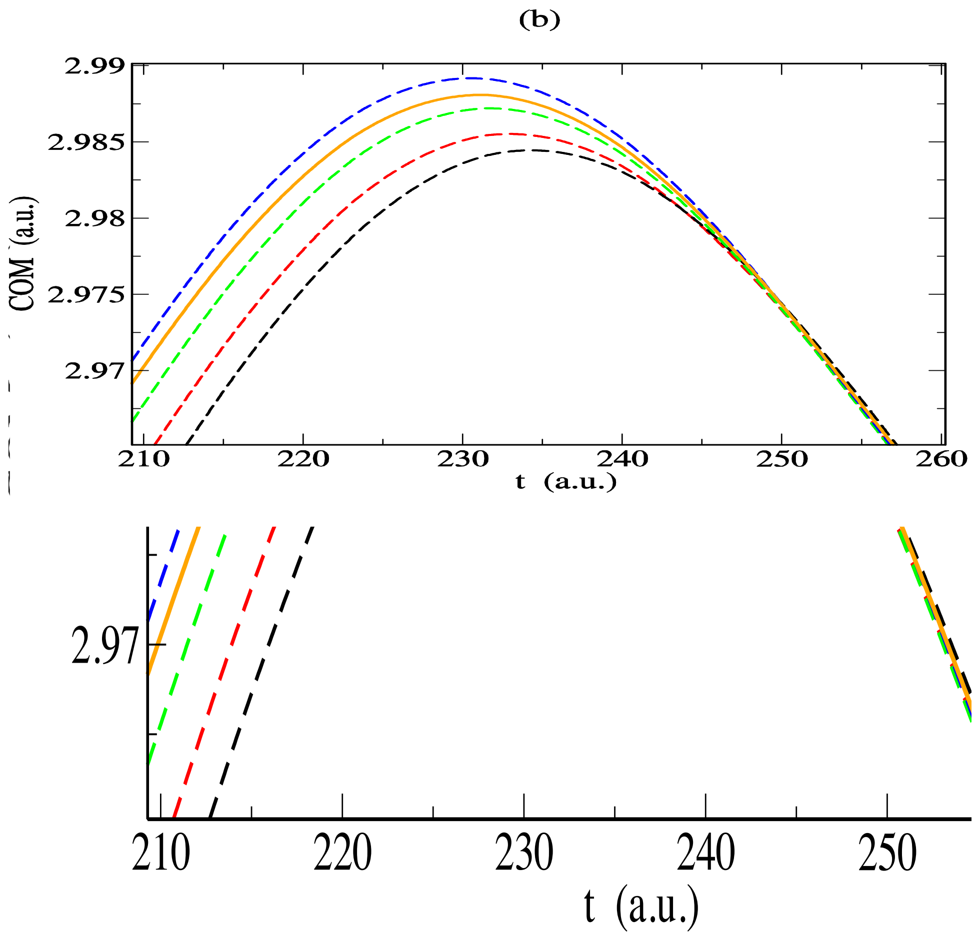
$$v(E = 3\text{ a.u.}) = 2.68 \pm 0.01\text{a.u.}$$
$$v(E = 2\text{ a.u.}) = 2.29 \pm 0.01\text{a.u.}$$

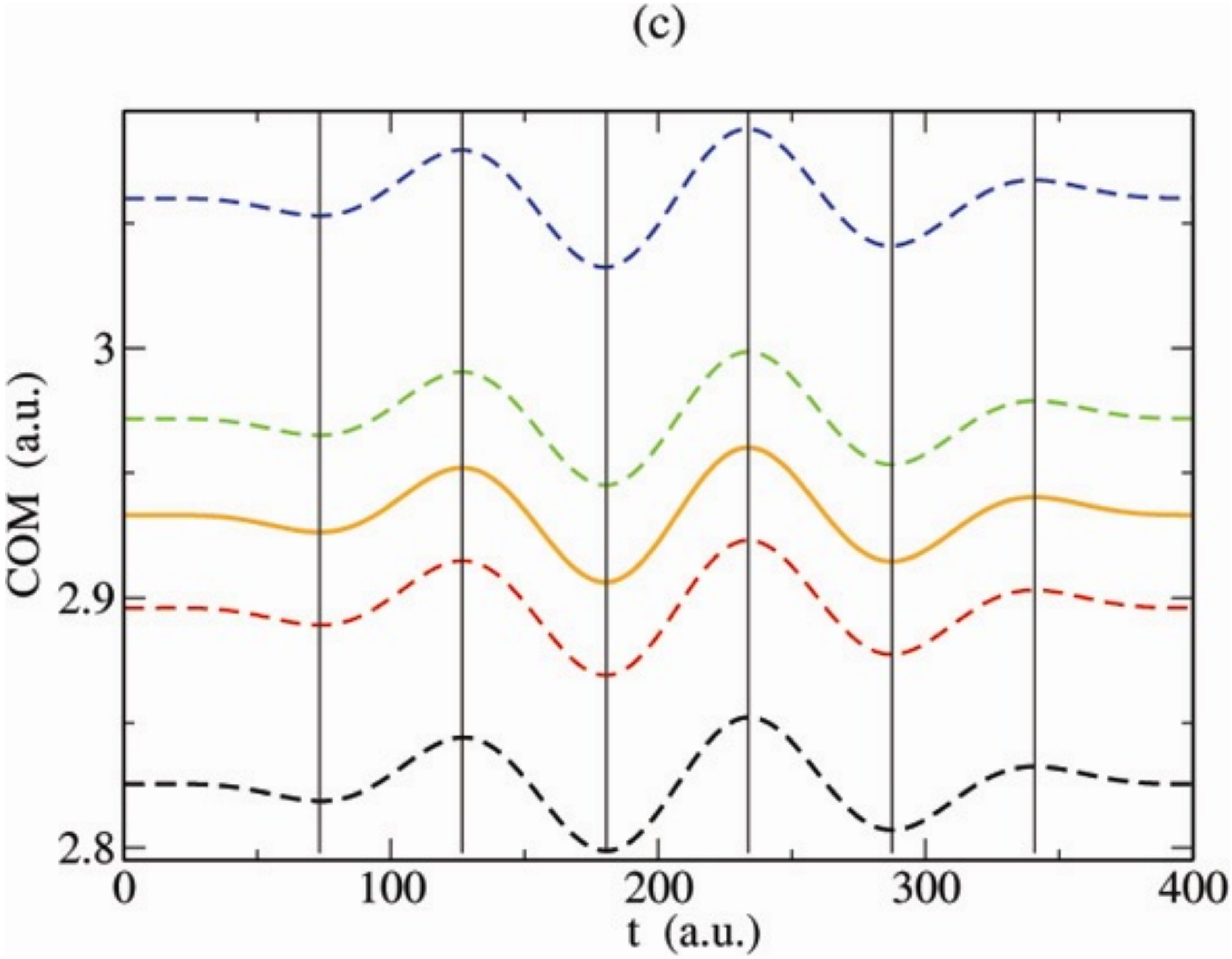
$$U_{av} \equiv E - v^2(E)\ /2$$

$$U_{av} = \begin{cases} -0.62\text{ a.u.} & E = 2\text{ a.u.} \\ -0.60\text{ a.u.} & E = 3\text{ a.u.} \end{cases}$$

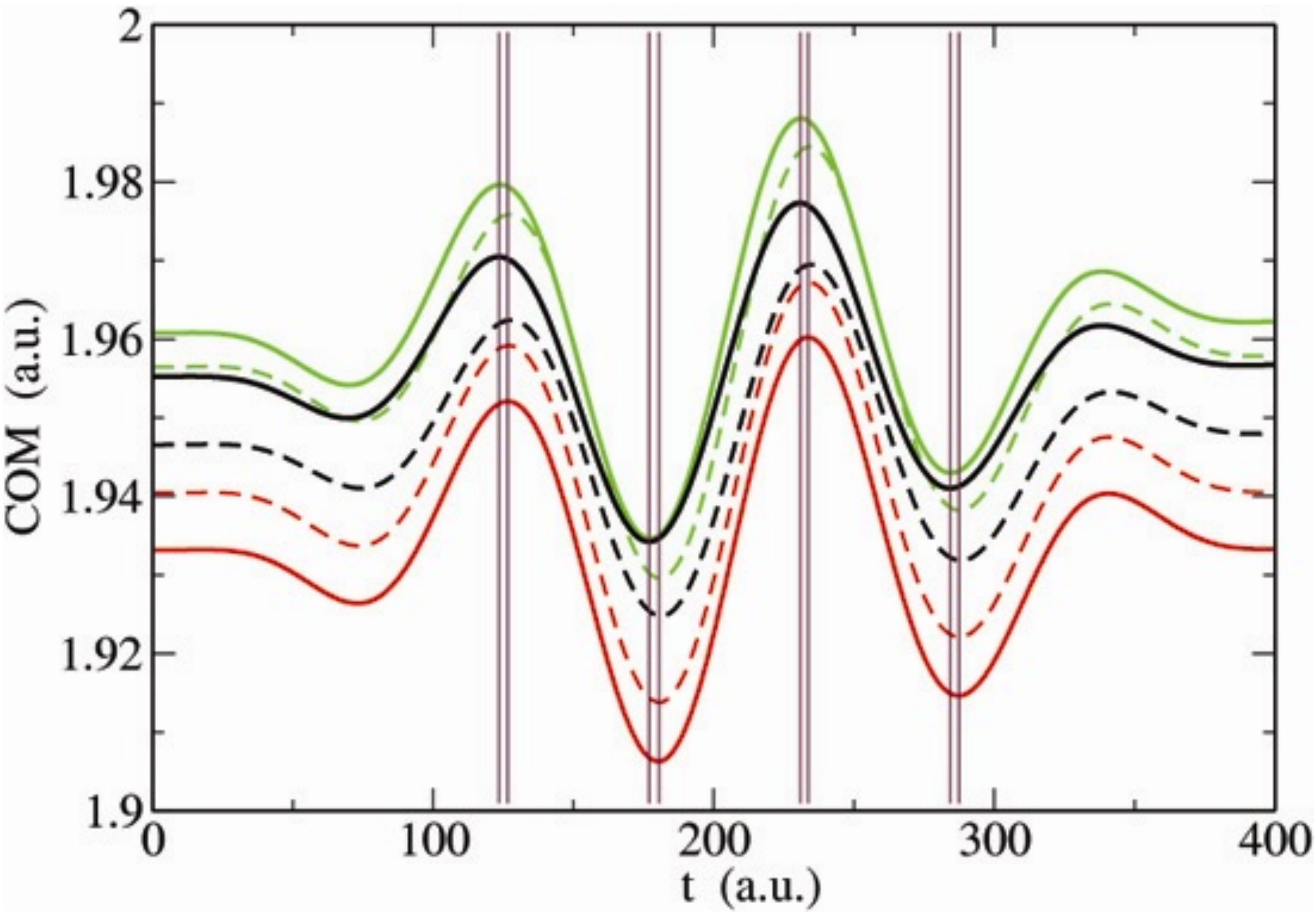
$$\varepsilon_F + A_{exit} \approx 12\text{ eV} = ...0.44\text{ a.u.}$$


(a)
1.98
1.975
1.97
1.965
1.96
1.955
COM (a.u.)
210
220
230
240
250
260
t (a.u.)
1.96
1.955
210
220
230
240
250
t (a.u.)

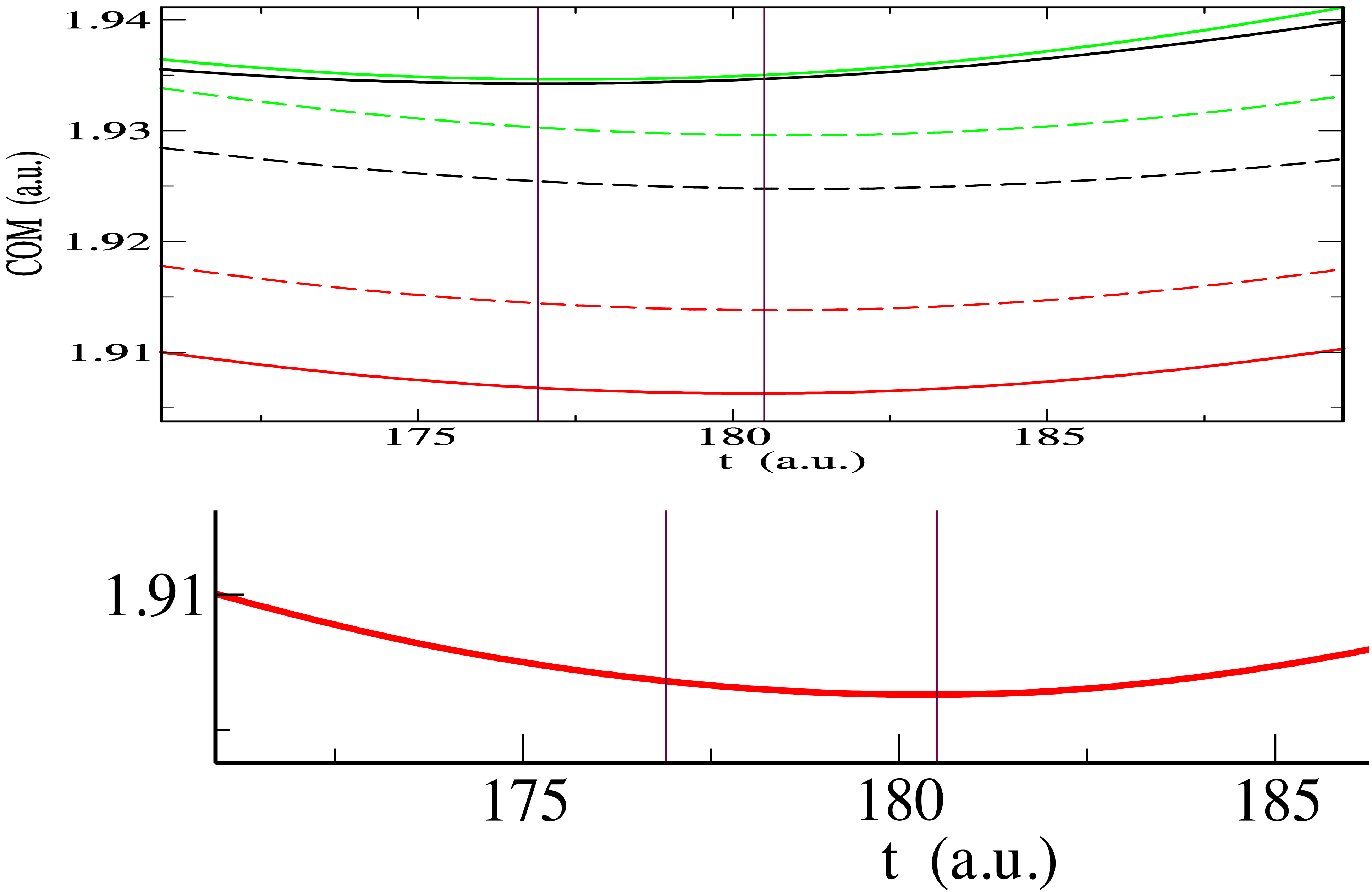

(b)
2.99
2.985
2.98
2.975
2.97
COM (a.u.)
210
220
230
240
250
260
t (a.u.)
2.97
210
220
230
240
250
t (a.u.)



(c)
3
2.9
2.8
COM (a.u.)
0
100
200
300
400
t (a.u.)



COM (a.u.)
t (a.u.)
2
1.98
1.96
1.94
1.92
1.9
0
100
200
300
400



1.94
1.93
1.92
1.91
COM (a.u.)
175
180
185
t (a.u.)
1.91
175
180
185
t (a.u.)

Result:

$$\Delta t_{theor} \cong 85\, as; \qquad \Delta t_{\exp} \cong 110 \pm 70\, as.$$

Conclusion:

The experimental result *can be* explained with interplay of two mechanisms:

- difference in the velocity of the ejected electrons in the final state in the bulk,
- dependence of the features on the character of the initial states: electron ejection from a localized *f*-state versus a delocalized *d*-state
- .

Principle questions:

- Can the relaxation processes be observed with attostreaking?

  1. Relaxation of the image potential in the case of photoionization of adsorbates.

  2. Dynamics of charge transfer from bulk to the adsorbate.

  3. Dynamics of screening of a hole in the bulk with observation of the Auger process.

- What can we try to compute in the tasks with simple adsorbates?

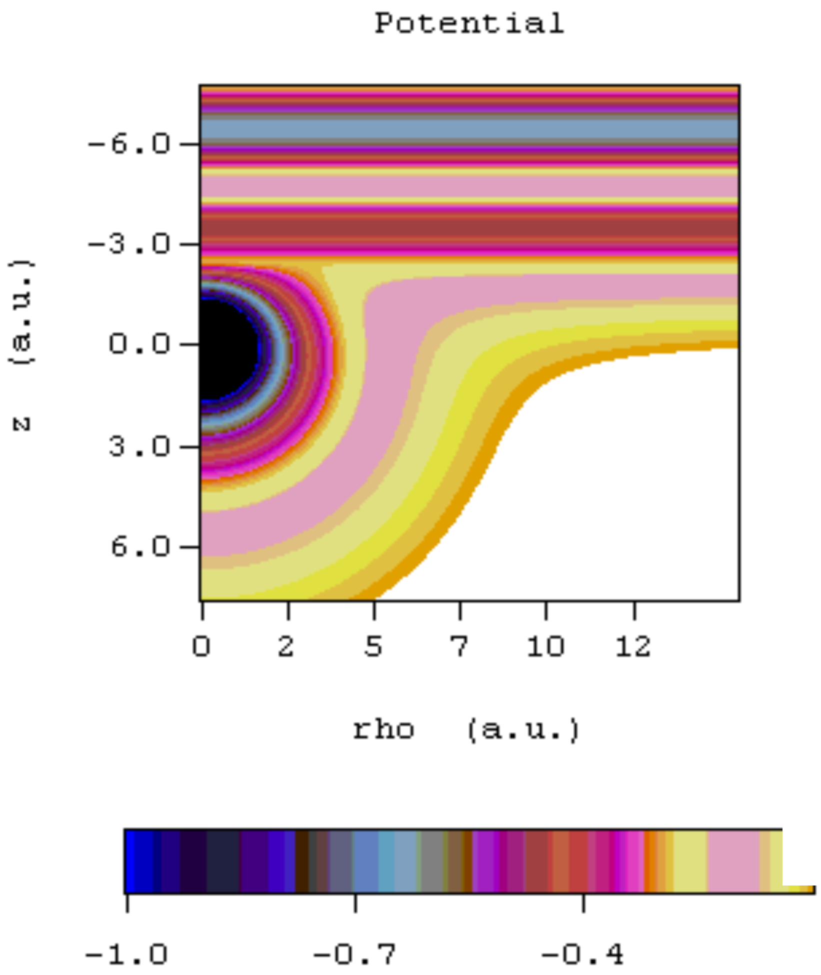
(A.K.Kazansky and P.M.Echenique, Phys.Rev.B 81,…, 2010)

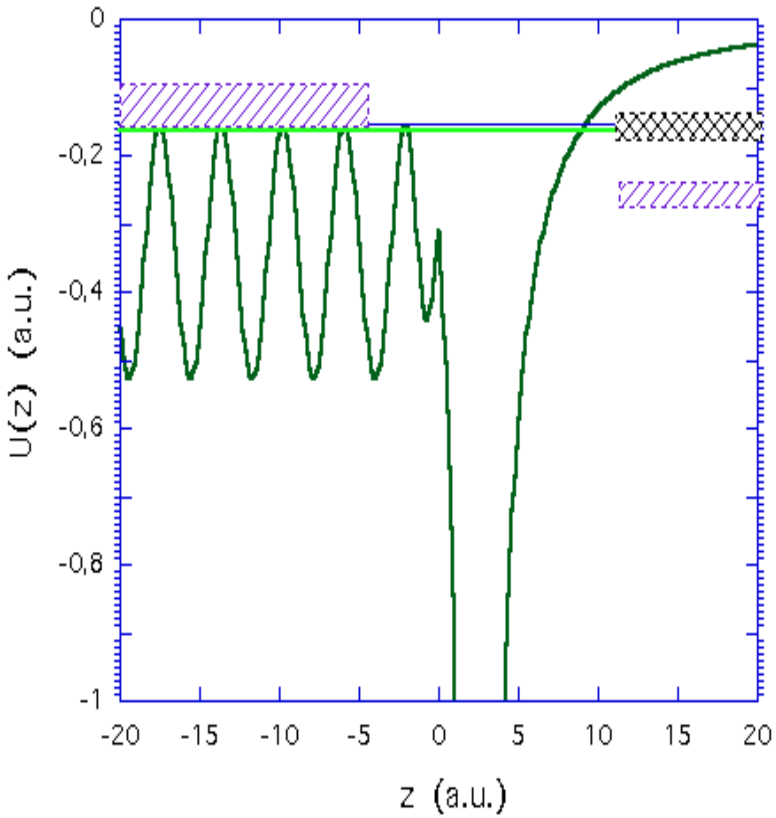
Basic model:

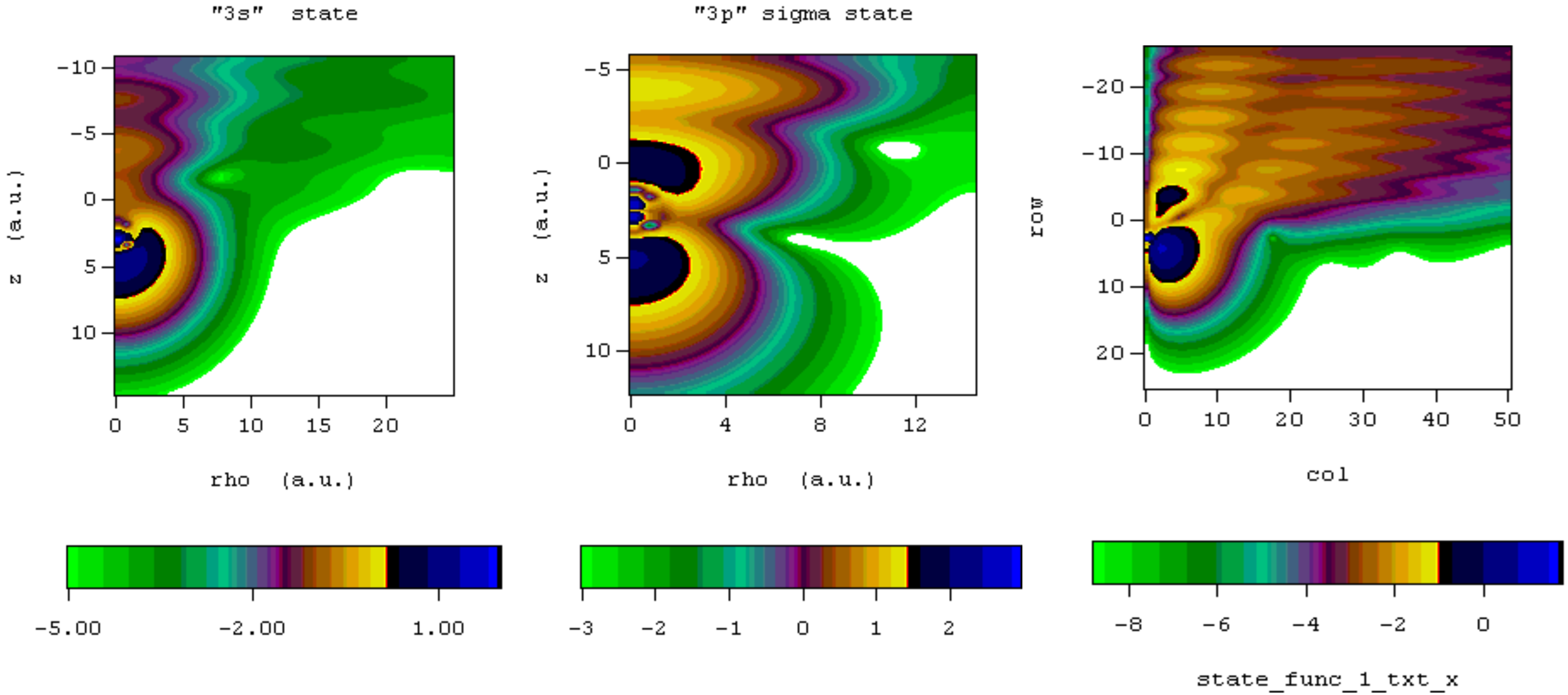
$$i\frac{\partial}{\partial t}\Psi(\vec{\rho},z;t) = -\frac{1}{2}\Delta\Psi(\vec{\rho},z;t) + U_2(\vec{\rho},z)\Psi(\vec{\rho},z;t)$$

$$+\left[U_{surf}(z) + U_{image}(\vec{\rho},z)\right]\Psi(\vec{\rho},z;t) - i\frac{\gamma_0(z)}{2}\Psi(\vec{\rho},z;t)$$

$$+\left[E_{IR}(t)f(z) + E_{XUV}(t)z\right]\Psi(\vec{\rho},z;t)$$


Potential
-6.0
-3.0
0.0
3.0
6.0
z (a.u.)
0
2
5
7
10
12
rho (a.u.)
-1.0
-0.7
-0.4


0
-0,2
-0,4
-0,6
-0,8
-1
U(z) (a.u.)
-20
-15
-10
-5
0
5
10
15
20
z (a.u.)



$$E_{'3p_\sigma'} = -0.128 - i0.034\ a.u.;$$

$$E_{'3p_\pi'} = -0.160 - i0.0023\ a.u.;$$

$$E_{'3s_\sigma'} = -0.264 - i0.0066\ a.u.;$$

$$E_{'2p_\sigma'} = -1.392\ a.u.\ (E_{2p}^{(0)} = -1.689\ a.u.)$$

$$E_{'2p_\pi'} = \qquad\qquad ;\ E_F = 4.43\ eV \quad (-0.162\ a.u.)$$

Charge exchange with the surface.

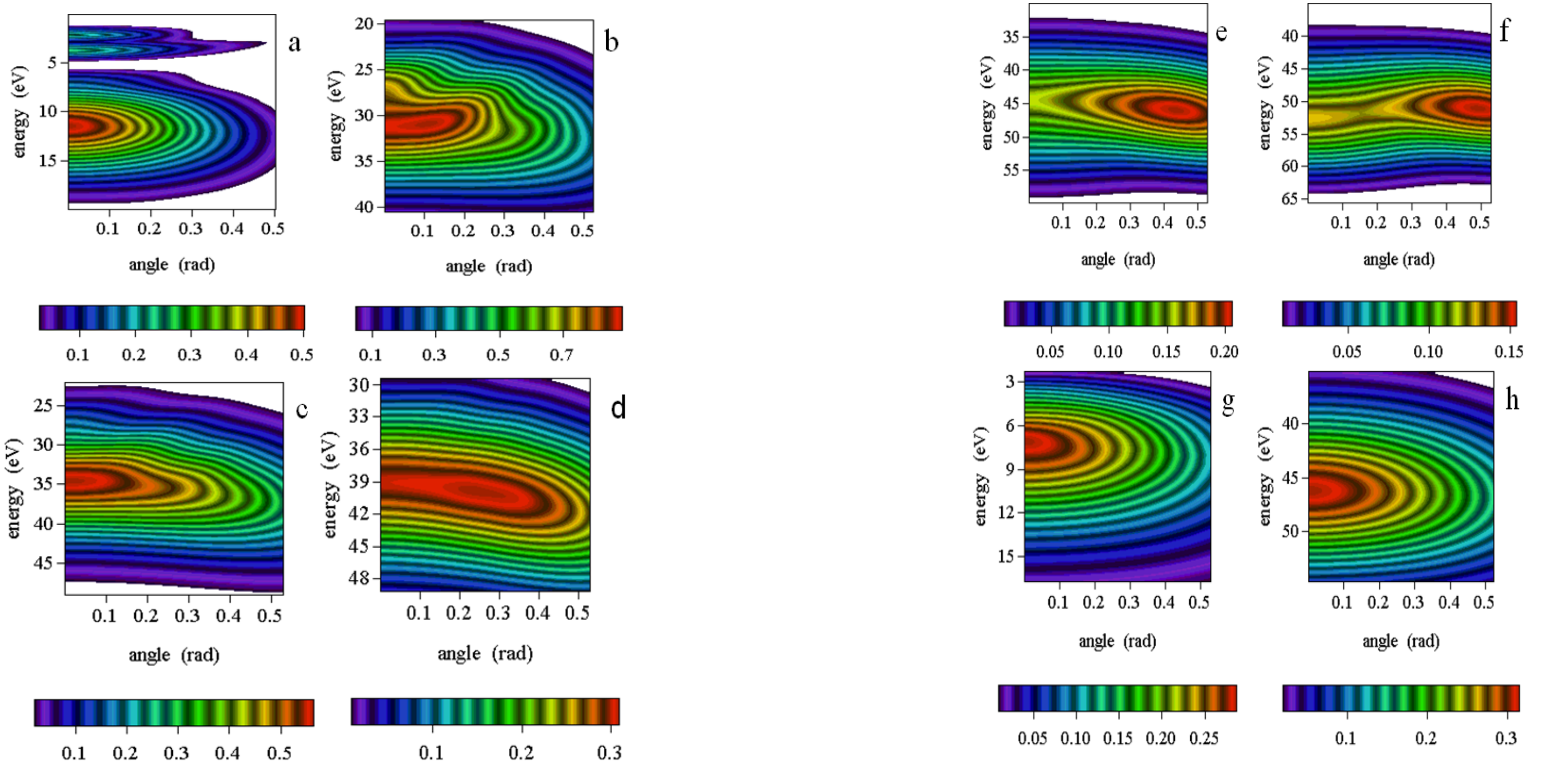
$$i\frac{\partial}{\partial t}\Psi_2(\bar{\rho},z;t) = -\frac{1}{2}\Delta\Psi_2(\bar{\rho},z;t) + U_2(\bar{\rho},z)\Psi_2(\bar{\rho},z;t)$$

$$+\left[U_{surf}(z) + U^{(2)}_{image}(\bar{\rho},z) - i\frac{\Gamma}{2}\right]\Psi_2(\bar{\rho},z;t).....;$$

$$i\frac{\partial}{\partial t}\Psi_1(\bar{\rho},z;t) = -\frac{1}{2}\Delta\Psi_1(\bar{\rho},z;t) + U_1(\bar{\rho},z)\Psi_1(\bar{\rho},z;t)$$

$$+\left[U_{surf}(z) + U^{(1)}_{image}(\bar{\rho},z)\right]\Psi_1(\bar{\rho},z;t).....+C\Psi_2(\bar{\rho},z;t).$$

$$C = ???? \quad - \quad C = i\frac{\Gamma}{2}\,?!$$


a
b
c
d
e
f
g
h
energy (eV)
angle (rad)
0.1 0.2 0.3 0.4 0.5
5
10
15
20
25
30
35
40
45
0.1 0.3 0.5 0.7
33
36
39
42
48
50
55
60
65
0.05 0.10 0.15 0.20
0.05 0.10 0.15
0.05 0.10 0.15 0.20 0.25
3
6
9
12
0.1 0.2 0.3

Dynamical screening and
formation of the image charge.

(A.K.Kazansky, P.M.Echenique, submitted)

$$\rho_e\left(\vec{\rho}, z, t\right) = -\delta\left(z - z_1(t)\right)\delta^2\left(\vec{\rho}\right)\theta(t)$$

$$-\delta\left(z - z_0\right)\delta^2\left(\vec{\rho}\right)\theta(-t);$$

$$\varphi_r\left(\vec{\rho}, z, t\right) = \int d^2\vec{K}\, d\omega\, dz'\, e^{i\left(\vec{K}\vec{\rho}\right) - i\omega t}$$

$$\overline{W}\left(\vec{K}, z, z', \omega\right)\rho_e\left(\vec{K}, z, \omega\right)$$

$$\overline{W}\left(\vec{K}, z, z', \omega\right) = \frac{e^{-K(z+z')}}{(2\pi)^{1/2} K} g(K, \omega);$$

$$g(K, \omega) = \frac{\varepsilon_s(K, \omega) - 1}{\varepsilon_s(K, \omega) + 1};$$

$$\varepsilon_s(K, \omega) = 1 - \frac{\omega_p^2}{\omega(\omega + i\gamma) - \omega_p^2 - \alpha K - \beta^2 K^2 - K^4/4}$$

$$\varepsilon_s(K, \omega_{sp}) + 1 = 0 \Rightarrow \omega_{sp}(K) = \frac{\omega_p}{\sqrt{2}} + aK + bK^2 - i\gamma/2$$

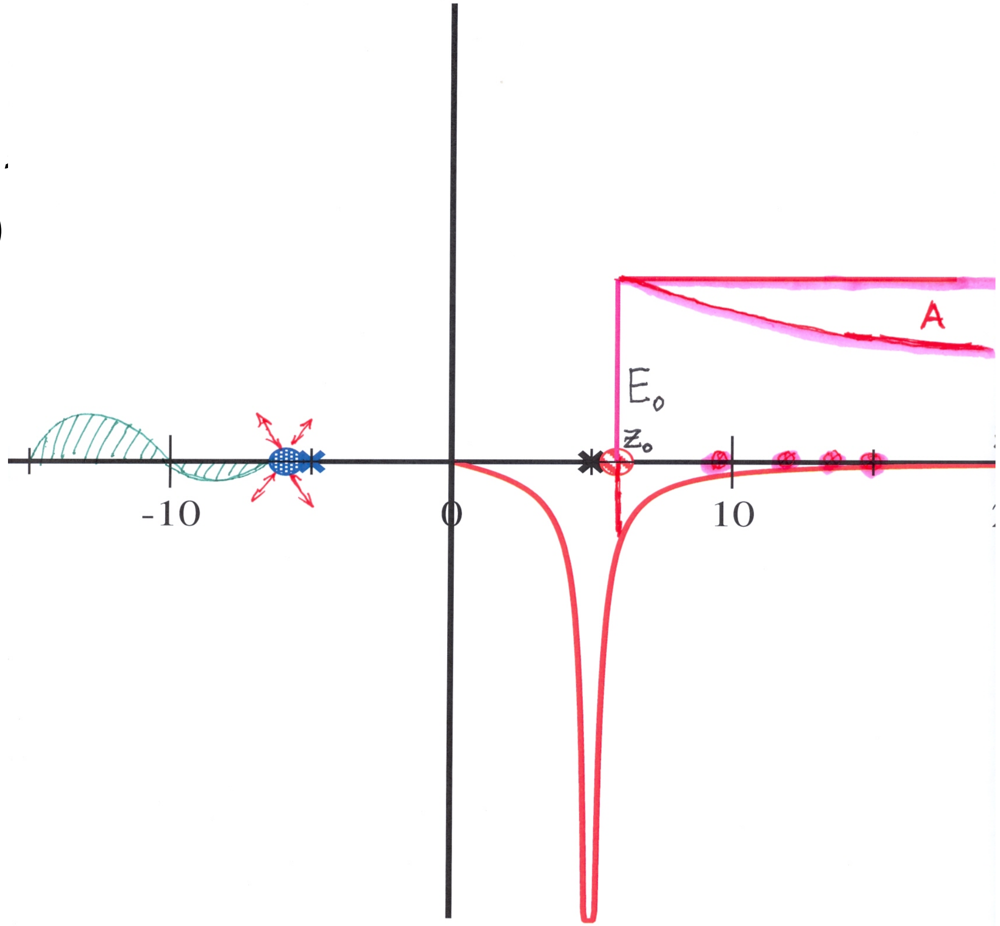
$$\frac{dz_1(t)}{dt} = \sqrt{2\left(E(t) - U_c(z_1(t)) - U_c^{imag}(z_1(t))\right)};$$

$$\frac{dE(t)}{dt} = v(t) F_r(z_1(t)),$$

$$F_r = \frac{\partial}{\partial z} \phi_r(z, t) \Big|_{z=z_1(t)}$$

$$z_1(0) = z_0,$$

$$E(0) = E_0.$$



$$F_{r1}(t) = -\int_0^{\infty} K\, dK\, e^{-K(z_1(t)+z_0)-\gamma t/2} \left[\cos(D(K)t) + \frac{\gamma \sin(D(K)t)}{2D(K)} \right],$$

$$F_{r2}(t) = \mathbf{Im}\left[\int_0^{\infty} K\, dK\, \frac{e^{-Kz_1(t)-\gamma t/2 - iD(K)t}}{D(K)} G(K,t) \right];$$

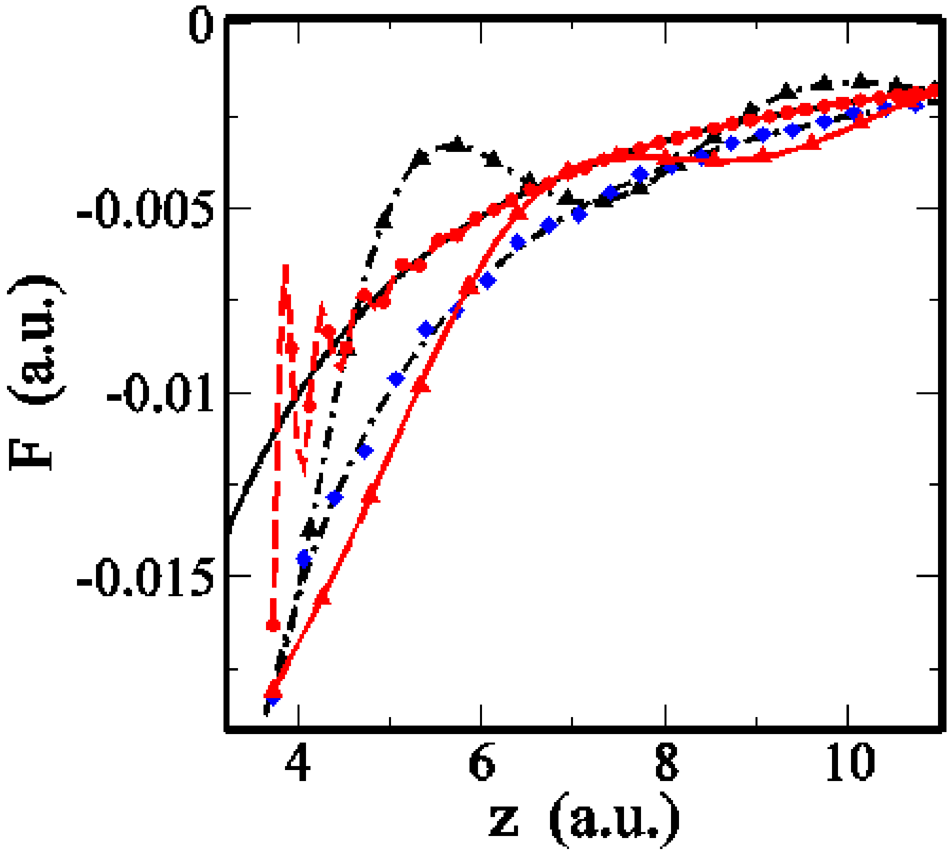
$$G(K,t) = \frac{\omega_p^2}{2} \int_0^t d\tau\, e^{-Kz(\tau)+\gamma\tau/2+iD(K)\tau};$$

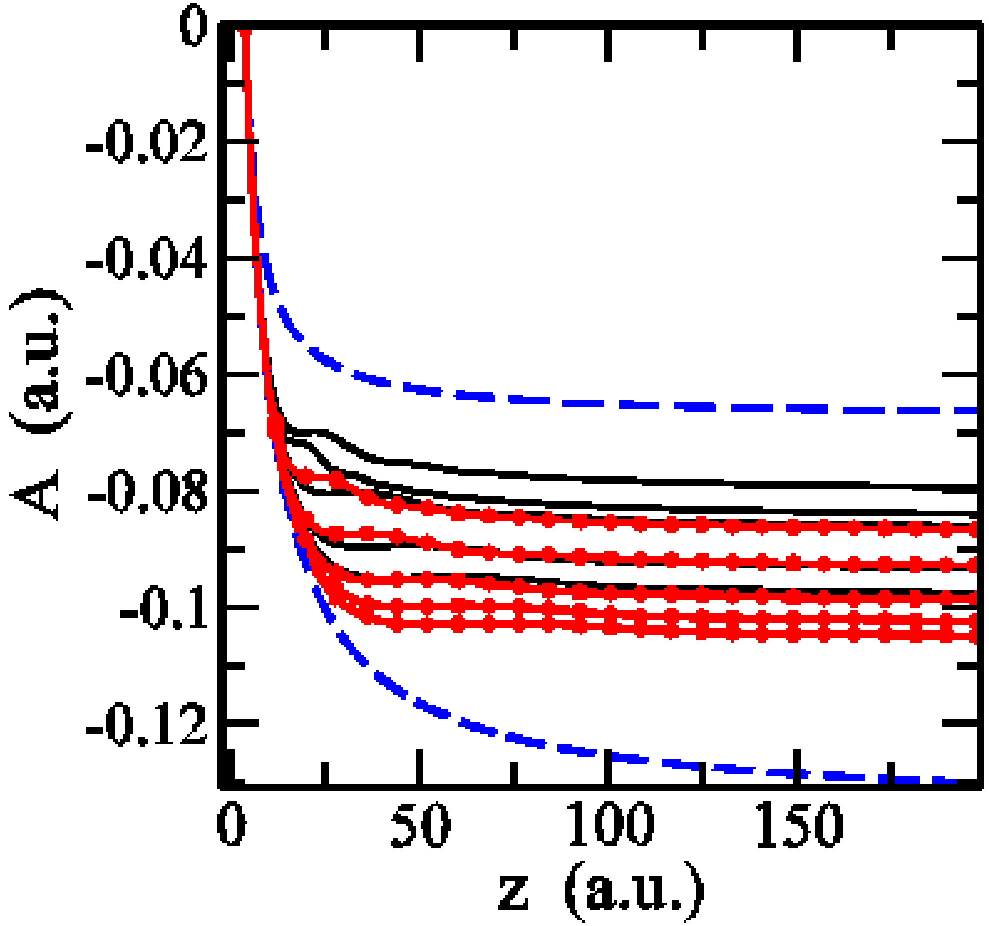
$$D(K) = \sqrt{-\frac{\gamma^2}{4} + \frac{\omega_p^2}{2} + \alpha K + \beta^2 K^2 + K^4/4}.$$

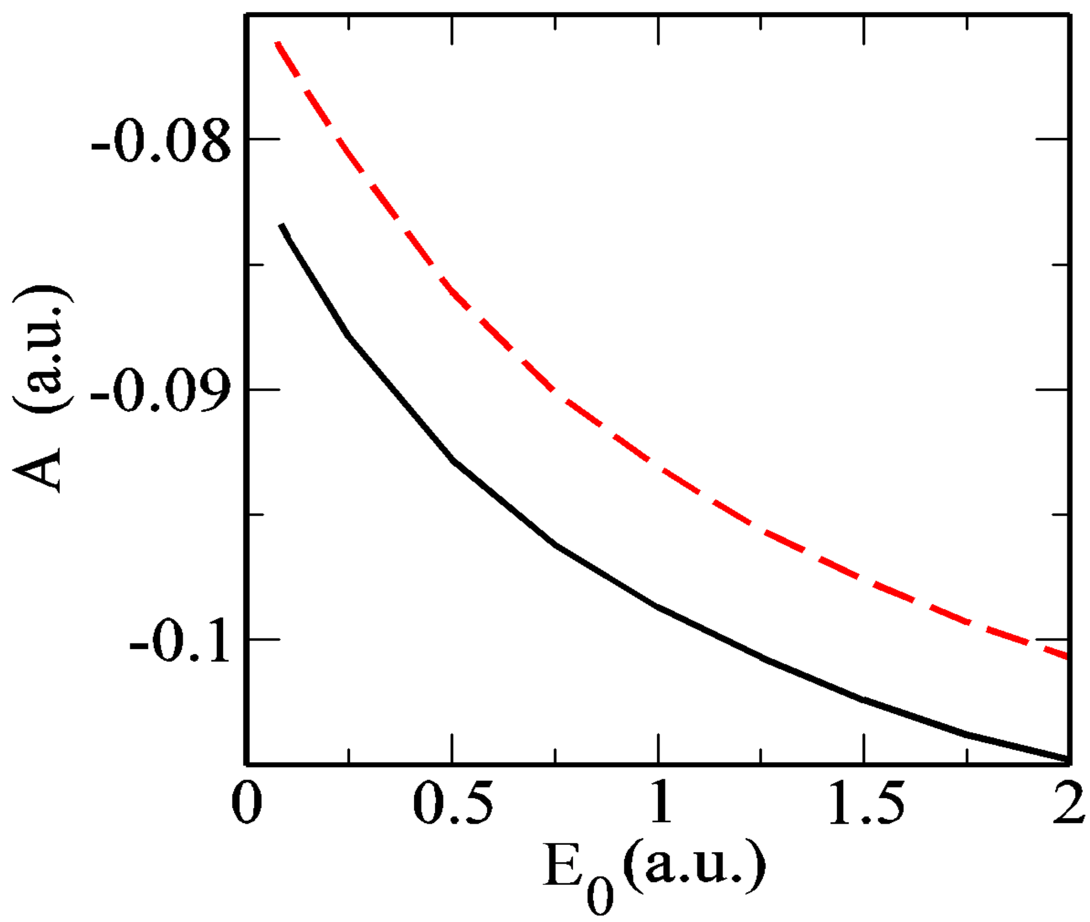
$$F_{r1}(t) = -\frac{e^{-\gamma\tau/2}}{\left(z_1(t)+z_0\right)^2}\left[\cos(Dt) + \frac{\gamma\sin(Dt)}{2D}\right],$$

$$F_{r2}(t) = -\frac{\omega_p^2}{2D}\int_0^t d\tau\,\frac{e^{-\gamma(t-\tau)/2}\sin\left(D(t-\tau)\right)}{\left(z_1(t)+z_1(\tau)\right)^2}.$$

$$F_{r2}(t) \cong \begin{cases} -\dfrac{1-e^{-\gamma t/2}\left(\cos(Dt)+\gamma/2D\sin(Dt)\right)}{2z_0^2}, & z_1(t)\cong z_0; \\ -\dfrac{1}{2z_1^2(t)}, & z_1(t) >> z_0 \end{cases}$$


0
-0.005
-0.01
-0.015
F (a.u.)
4
6
8
10
z (a.u.)


0
-0.02
-0.04
-0.06
-0.08
-0.1
-0.12
A (a.u.)
0
50
100
150
z (a.u.)



-0.08
-0.09
-0.1
A (a.u.)
0
0.5
1
1.5
2
E0 (a.u.)