Physics of surfaces and interfaces

3. Scanning tunneling microscopy: quasiparticle interference and quantum corrals. Quantum-well states and tunneling interferometry

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Outline of current lecture

* Basic principles of scanning tunneling microscopy and spectroscopy
* Bardeen's approach for calculation of tunneling current
* Theory of STM: approach of Tersoff and Hamann
* Quasiparticle interference and quantum corrals
* Quantum-well states and tunneling interferometry
* Stark-shifted image-potential states and field-emission resonances

Part I

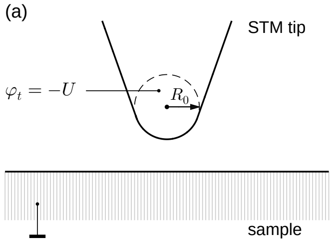
Theory of STM: approach of Tersoff and Hamann

(continuation)

Problem of Tersoff and Hamann

Tersoff and Hamann, Phys. Rev. Lett. vol. 50, 1998 (1983)

We consider tunneling effect between metallic semi-space with flat surface (at $z < 0$) and metallic tip with an apex of spherical shape of the radius R. For sake of simplicity we assume that the Fermi energies and the work functions are equal: $E_F^{(s)} = E_F^{(t)} = E_F$ and $W_s = W_t = W$.



Here U is the potential of the sample with respect to a tip ($\varphi_s = 0$ and $\varphi_t = -U$).

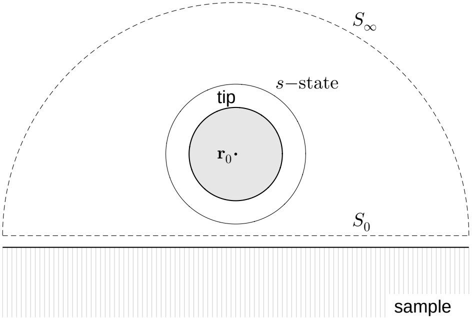
Let $T_{s\to t}$ be the matrix element, corresponding to the transition of the electron from one of possible states in the sample with wave function ψ_s and energy E_s to one of possible states in the tip with wave function ψ_t and energy E_t.

Effect of $s-$orbital to the tunneling conductance (1)

We consider a surface S_0 lying between the sample and the tip and a surface of infinite radius S_∞. Since both ψ_s and ψ_t are exponentially decay at the increase of the distance from the corresponding surfaces, the surface integral over то S_∞ is equal to zero

$$T^{(s)}_{s\to t} = -\frac{\hbar^2}{2m^*} \int\limits_{S_0+S_\infty} \left\{ \psi_t^{(s)} \frac{\partial \psi_s}{\partial n} - \psi_s \frac{\partial \psi_t^{(s)}}{\partial n} \right\} dS =$$

$$= -\frac{\hbar^2}{2m} \iiint\limits_{z>0} \left\{ \psi_t^{(s)} \nabla^2 \psi_s - \psi_s \nabla^2 \psi_t^{(s)} \right\} dV.$$



Effect of $s-$orbital to the tunneling conductance (2)

Taking into account that $\nabla^2\psi_s = \varkappa^2\psi_s$, $\nabla^2\psi_t^{(s)} = \varkappa^2\psi_t^{(s)} - 4\pi C\varkappa^{-1}\,\delta(\boldsymbol{r}-\boldsymbol{r}_0)$, and $\psi_t^{(s)\,*} = \psi_t^{(s)}$, we come to the important result

$$T^{(s)}_{s\to t} = -\frac{\hbar^2}{2m^*}\int\limits_{z>0}\left\{\psi_t^{(s)}\,\varkappa^2\psi_s - \psi_s\,\varkappa^2\psi_t^{(s)} + \psi_s\,\frac{4\pi C}{\varkappa}\,\delta(\boldsymbol{r}-\boldsymbol{r}_0)\right\}dV =$$

$$= -\frac{\hbar^2}{2m^*}\int\limits_{z>0}\psi_s(\boldsymbol{r})\,\frac{4\pi C}{\varkappa}\,\delta(\boldsymbol{r}-\boldsymbol{r}_0)\,dV = -\frac{\hbar^2}{2m^*}\,\frac{4\pi C}{\varkappa}\cdot\psi_s(\boldsymbol{r}_0).$$

Intermediate conclusion: the matrix element of the tunneling transition is proportional to the wave function of the electron of the sample in the center of the tip provided that the dominant contribution is associated with the $s-$wave orbitals of the electrons near the tip apex.

As a result, the square of the absolute value of the matrix element is proportional to the probability to detect the electron of the sample in the center of the tip

$$\left|T^{(s)}_{s\to t}\right|^2 = \left(\frac{\hbar^2}{2m^*}\right)^2\left(\frac{4\pi}{\varkappa}\right)^2|C|^2\cdot|\psi_s(\boldsymbol{r}_0)|^2\,.$$

Effect of $s-$orbital to the tunneling conductance (3)

After summation we get the differential tunneling conductance

$$\left(\frac{dI}{dU}\right)^{(s)} = \frac{2\pi e^2}{\hbar}\left(\frac{\hbar^2}{2m^*}\right)^2\left(\frac{4\pi}{\varkappa}\right)^2 |C|^2 \times$$
$$\times\left(\sum_s \left|\psi_s(\boldsymbol{r}_0)\right|^2 \delta\big(E_s - (E_F + |e|U)\big)\right)\cdot\left(\sum_t \big(E_t - (E_F + |e|U)\big)\right).$$

We define the integral density of states (DOS) for electrons in the tip with energy E (without spin degeneracy) as follows

$$\rho_t(E) = \sum_t 1\cdot\delta(E_t - E).$$

We define the local density of states (LDOS) for electrons in the sample with energy E (without spin degeneracy) as follows

$$\rho_s(\boldsymbol{r}_0, E) = \sum_s \left|\psi_s(\boldsymbol{r}_0)\right|^2 \delta(E_s - E).$$

Thus, the differential tunneling conductance in this model is equal to

$$\left(\frac{dI}{dU}\right)^{s-wave} \simeq const\cdot\rho_s(\boldsymbol{r}_0, E_F + |e|U)\cdot\rho_t(E_F).$$

Modeling of electronic structure of surface (1)

Tersoff and Hamann, Phys. Rev. Lett. vol. 50, 1998 (1983);
Tersoff and Hamann, Phys. Rev. B, vol. 31, 810 (1985).

$$\rho_s(\boldsymbol{r}_0, E_F + |e|U) = const$$

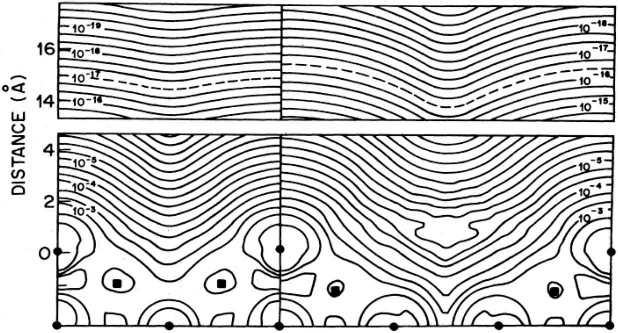


FIG. 2. Calculated $\rho(\vec{r}, E_F)$ for Au(110) (2×1) (left) and (3×1) (right) surfaces. Figure shows $(1\bar{1}0)$ plane through outermost atoms. Positions of nuclei are indicated by solid circles (in plane) and squares (out of plane). Contours of constant $\rho(\vec{r}, E_F)$ are labeled in units of a.u.$^{-3}$eV^{-1}. Note break in distance scale. Peculiar structure around contour 10^{-5} of (3×1) is due to limitations of the plane-wave part of the basis in describing the exponential decay inside the deep troughs. Center of curvature of probe tip follows dashed line.

Modeling of electronic structure of surface (2)

Tupchaya, Bondarenko, Yakovlev, Vekovshinin, Mihalyuk, Gruznev, Denisov, Matetskiy, Aladyshkin, Zotov, Saranin, Applied Surface Science, vol. 589, 152951 (2022).

$$\rho_s(\boldsymbol{r}_0, E_F + |e|U) = const$$

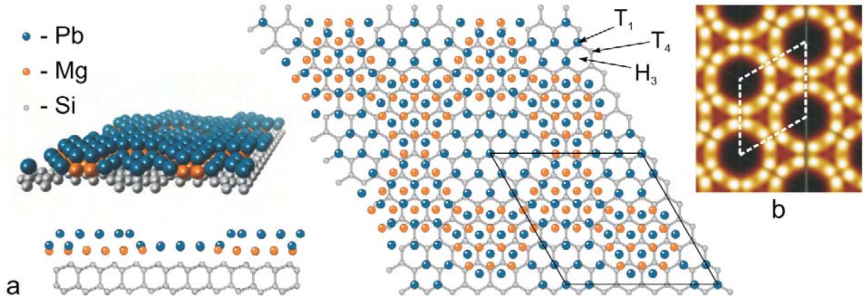
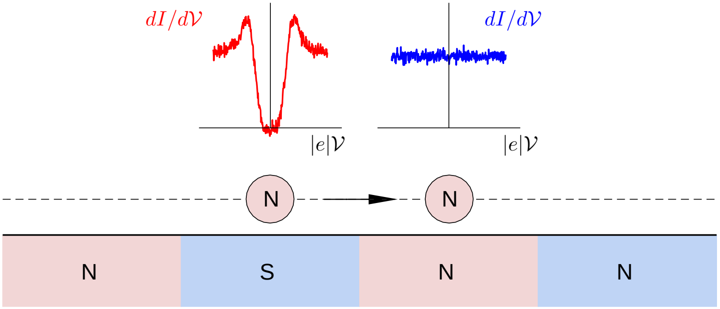


Fig. 4. (a) Ball-and-stick (top and side views) and isometric view of the structural model for the 6 × 6-(Mg, Pb) surface. (b) Simulated empty state STM image (+0.5 V).

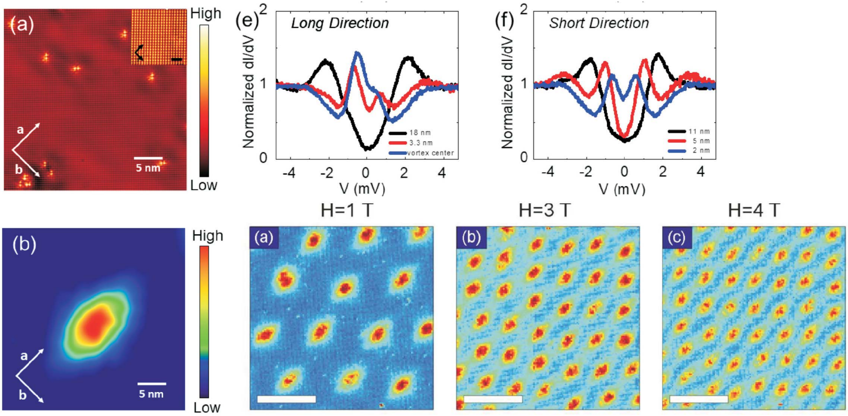
Scanning tunneling spectroscopy and physics of superconductivity (1)

Scanning tunneling microscopy makes possible to detect non-superconducting inclusions in superconducting environment and, in particular, normal cores of Abrikosov vortices:



Scanning tunneling spectroscopy and detection of vortices (1)

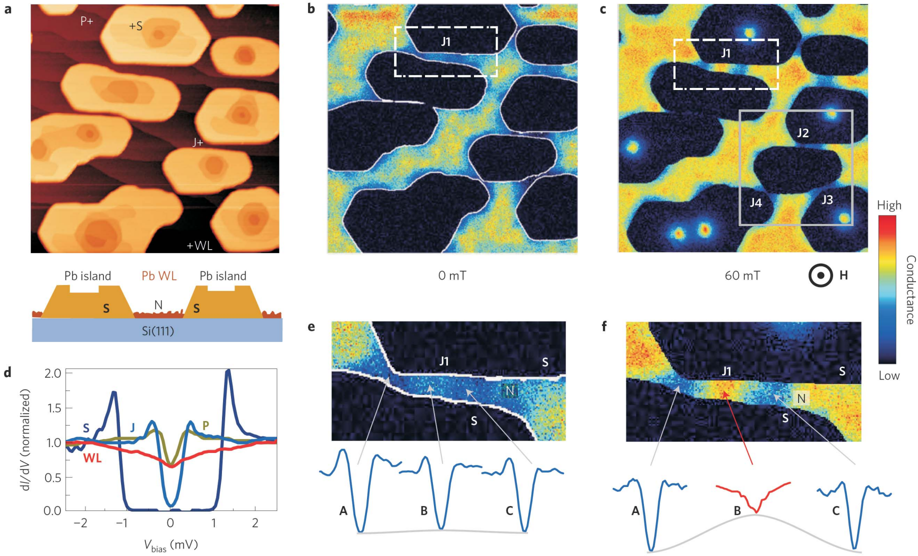
Lattice of Abrikosov vortices in superconducting FeSe single crystals



Putilov, Di Giorgio, Vadimov, Trainer, ... Aladyshkin, Mel'nikov, Iavarone. Phys. Rev. B, vol. 99, 144514 (2019)

Scanning tunneling spectroscopy and detection of vortices (2)

Individual Abrikosov vortices in superconducting Pb(111) micron-sized islands

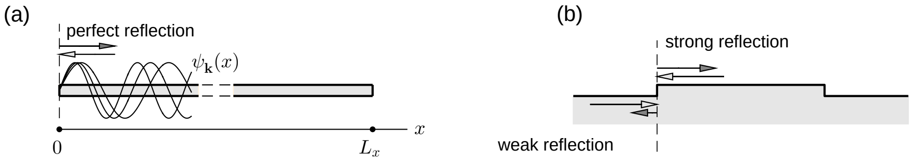


Roditchev, Brun, Serrier-Garcia, Cuevas, ..., Stolyarov, Nature Physics, vol. 11, 322-337 (2015)

Part II

Quasiparticle interference and quantum corrals

Quasiparticle interference near defects at surface (1)



We consider two-dimensional electronic gas either near the flat edge at $x = 0$ (panel a) or surface electronic waves at surface with a single monatomic step at $x = 0$ (panel b). In both cases the normalized wave functions near the edge have the following form

$$\Psi_{\boldsymbol{k}}(x, y) = \sqrt{\frac{2}{L_x L_y}} \cdot \sin k_x x \cdot e^{ik_y y} \quad \text{and} \quad |\Psi_{\boldsymbol{k}}(x, y)|^2 = \frac{2}{L_x L_y} \cdot \sin^2 k_x x.$$

Assume that the energy dispersion is isotropic

$$E_{\boldsymbol{k}} = f(k), \quad \text{where} \quad k = \sqrt{k_x^2 + k_y^2}.$$

It covers two cases of both massive particle and mass-less electrons

$$E_{\boldsymbol{k}} = E_0 + \frac{\hbar^2 k^2}{2m^*} \quad \text{and} \quad E_{\boldsymbol{k}} = E_0 + \alpha \cdot k.$$

Quasiparticle interference near defects at surface (2)

According to the definition, the local density of states accounting spin degeneracy is equal

$$\rho(E,x,y) = 2 \cdot \sum_{\boldsymbol{k}} |\psi_s(\boldsymbol{r}_0)|^2 \cdot \delta(E_{\boldsymbol{k}} - E) = \frac{4}{L_x L_y} \cdot \sum_{\boldsymbol{k}} \sin^2 k_x x \cdot \delta(E_{\boldsymbol{k}} - E) =$$

$$= \frac{4}{L_x L_y} \cdot \frac{L_x}{2\pi} \frac{L_y}{2\pi} \cdot \iint \sin^2 k_x x \cdot \delta(E_{\mathrm{k}} - E) \cdot dk_x dk_y .$$

The integrand diverges at $|k| = k_0$, where k_0 is a positive root of the equation $E_{\boldsymbol{k}} - E = 0$. By expanding the function $E_{\boldsymbol{k}}$ in the Taylor series at $k \simeq k_0$

$$E_{\boldsymbol{k}} - E = \left(\frac{dE_{\boldsymbol{k}}}{dk} \right)_{k=k_0} \cdot (k - k_0),$$

we arrive at the following presentation of the delta-function of composed argument

$$\delta(E_{\boldsymbol{k}} - E) = \delta \left[\left(\frac{dE_{\boldsymbol{k}}}{dk} \right)_{k=k_0} \cdot (k - k_0) \right] = \left(\frac{dE_{\boldsymbol{k}}}{dk} \right)^{-1}_{k=k_0} \cdot \delta(k - k_0).$$

As a result,

$$\rho(E,x,y) = \frac{1}{\pi^2} \left(\frac{dE_{\boldsymbol{k}}}{dk} \right)^{-1}_{k=k_0} \cdot \iint \sin^2 k_x x \cdot \delta(k - k_0)\, dk_x dk_y .$$

Quasiparticle interference near defects at surface (3)

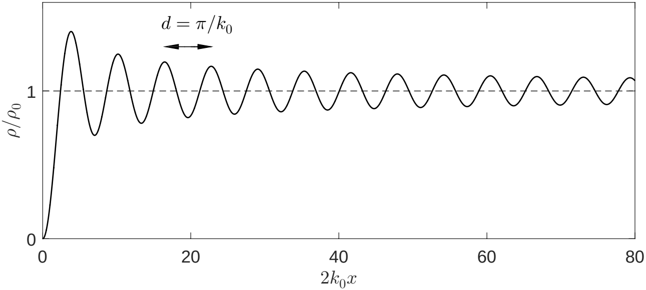
After integration in the cylindrical coordinate system, we get

$$\rho(\boldsymbol{r}, E) = \frac{1}{\pi^2}\left(\frac{dE_k}{dk}\right)^{-1}_{k=k_0} \cdot \iint \sin^2\left(kx\cos\theta\right)\cdot\delta(k-k_0)\cdot k\,dk\,d\theta =$$

$$= \frac{1}{\pi^2}\left(\frac{dE_k}{dk}\right)^{-1}_{k=k_0} \cdot k_0 \cdot \frac{1}{2}\left\{\int_{-\pi}^{\pi} d\theta - \int_{-\pi}^{\pi}\cos\left(2k_0 x\cos\theta\right)d\theta\right\} =$$

$$= \frac{1}{\pi}\left(\frac{dE_k}{dk}\right)^{-1}_{k=k_0} \cdot k_0 \cdot \left\{1 - J_0(2k_0x)\right\},$$

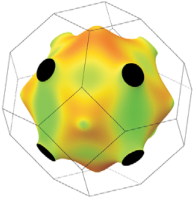
where $J_0(x)$ is the Bessel function of zero kind.

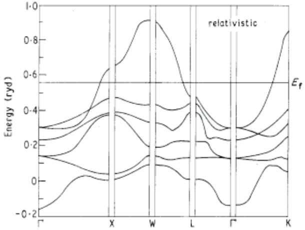


Quasiparticle interference for noble metals Au(111), Ag(111) и Cu(111)

Crommie, Lutz, Eigler, Nature, vol. 363, 524-527 (1993)

Crommie, Lutz, Eigler, Heller, Surf. Rev. Lett., vol. 2, 127-137 (1995)





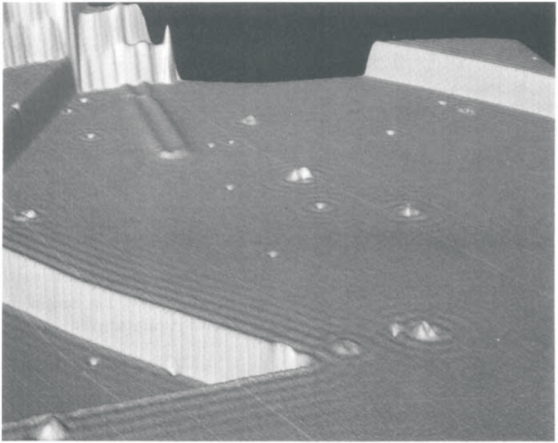


Fig. 1. Constant current STM image of the Cu(111) surface (V = 0.1 V, I = 1.0 nA). Spatial oscillations with a periodicity of 15 Å are clearly evident emanating from monatomic step edges and point defects. The vertical scale has been exaggerated to display the spatial oscillations more clearly.

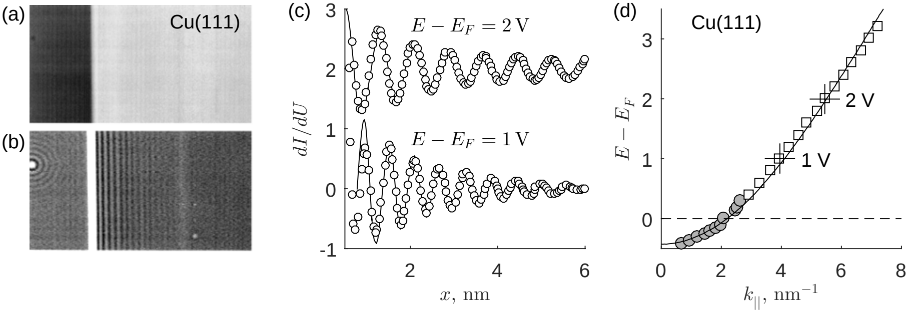
Dispersion of electronic surface states (1)

Quasiparticle interference and dephasing processes ($L_\varphi \neq \infty$)

$$\rho(E,x) = \rho_{bulk} + \frac{m^*}{\pi\hbar^2}\left\{1 - r(k_0)\, e^{-2x/L_\varphi}\, J_0(2k_0x)\right\}$$

$$\Longrightarrow \quad k_0 = k_0(E_F + |e|U) \quad \Longrightarrow \quad E - E_F = E_{||}(k_0).$$

Example: quasiparabolic spectrum of the surface states in noble metals

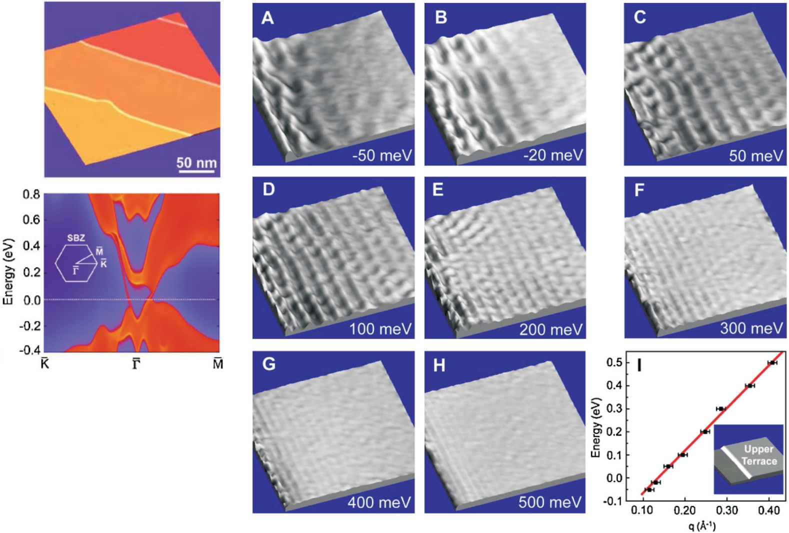


Crommie, Lutz, Eigler, Nature, vol. 363, 524-527 (1993)

Bürgi, Jeandupeux, Brune, Kern, Phys. Rev. Lett., vol. 82, 4516-4519 (1999)

Dispersion of electronic surface states (2)

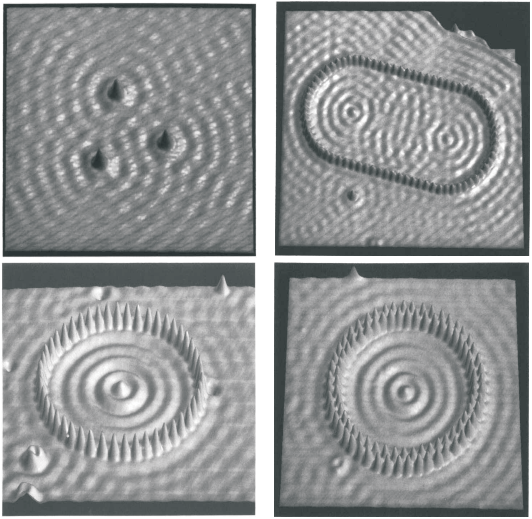
Example: 3D topological insulator Bi_2Te_3



Zhang et al., Phys. Rev. Lett., vol. 103, 266803 (2009)

Quasiparticle interference and quantum corrals

Adatoms Fe on atomically flat Cu(111) surface



Crommie, Lutz, Eigler, Heller, Surf. Rev. Lett., vol. 2, 127-137 (1995)

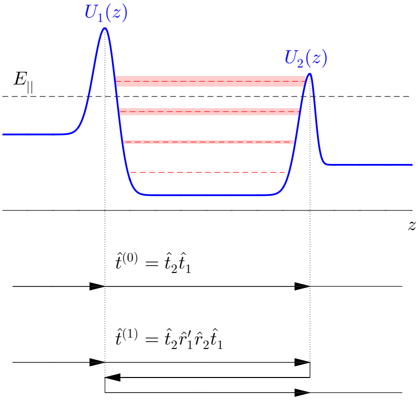
Part III

Quantum-well states and tunneling interferometry

Coherent resonant tunneling in a double-barrier structure (1)

Assume that the the potential $U(z)$ can be considered as a combination of two localized scatters (barriers or steps) $U_1(z)$ and $U_2(z)$. Assume that we know all amplitudes of reflection and transmission for these scatters. How to calculate the resulting coefficient of transmission?

We apply a perturbation theory, formally assuming that all reflection coefficients are rather small, while all transmission coefficients are close to unity.



zero-order term: $t^{(0)} = t_2\, t_1$

first-order term: $t^{(1)} = t_2\, r'_1\, r_2\, t_1$

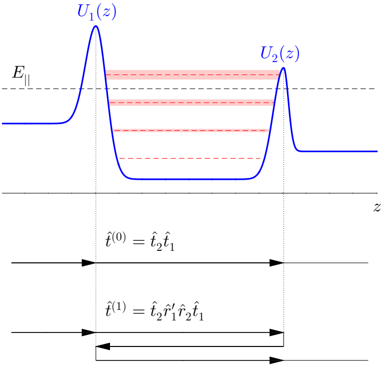
$n-$th order term: $t^{(n)} = t_2\, (r'_1\, r_2)^n\, t_1$

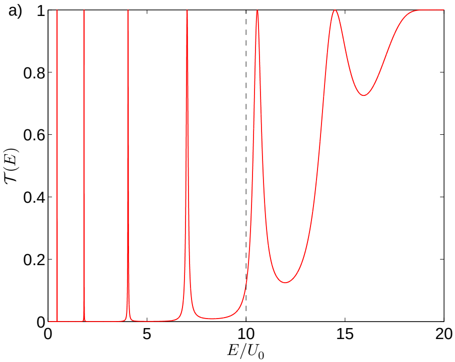
$$t = \sum_{n=0}^{\infty} t^{(n)} = \sum_{n=0}^{\infty} t_2\, \left(r'_1 r_2\right)^n\, t_1 = \frac{t_2\, t_1}{\left(1 - r'_1 r_2\right)},$$

provided that $\quad 1 - r'_1 r_2 \neq 0$

The conditions $1 - r'_1 r_2 = 0$ and $arg\ r'_1 + arg\ r_2 = 2\pi n$ correspond to the localized stationary or quasi-stationary electronic states in a quantum well.

Coherent resonant tunneling in a double-barrier structure (2)





$$t = \frac{t_1 t_2}{1 - r_1' r_2} \quad \text{and} \quad \mathcal{T} = \left| \frac{t_1 t_2}{1 - r_1' r_2} \right|^2 = \frac{\mathcal{T}_1 \mathcal{T}_2}{1 + \mathcal{R}_1 \mathcal{R}_2 - 2\sqrt{\mathcal{R}_1 \mathcal{R}_2} \cos\left(\arg r_1' + \arg r_2\right)},$$

where $\mathcal{R}_{1,2}$ and $\mathcal{T}_{1,2}$ are coefficients of reflection and transmission through the first and second barriers, correspondingly.

Coherent resonant tunneling in a double-barrier structure (3)

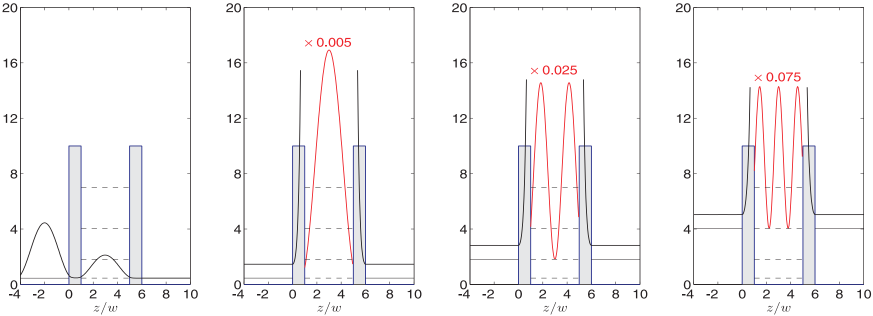
Transmission coefficient through double-barrier structure

$$\mathcal{T} = \left| \frac{t_1 t_2}{1 - r_1' r_2} \right|^2 = \frac{\mathcal{T}_1 \mathcal{T}_2}{1 + \mathcal{R}_1 \mathcal{R}_2 - 2\sqrt{\mathcal{R}_1 \mathcal{R}_2} \cos\left(\arg r_1' + \arg r_2\right)}.$$

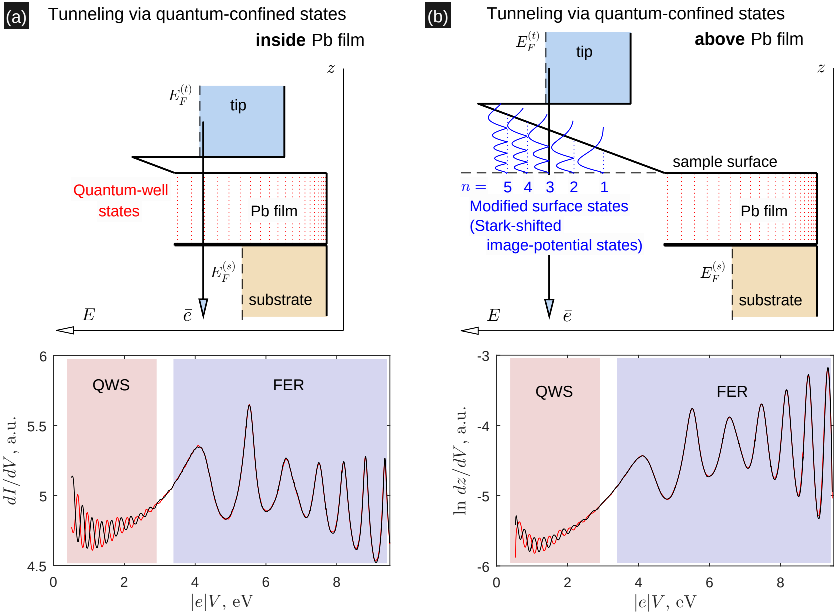
Transmission through low-transmission barriers ($\mathcal{T}_1 \ll 1$ and $\mathcal{T}_2 \ll 1$)

$$\mathcal{T}_{max} \simeq \frac{4\,\mathcal{T}_1 \mathcal{T}_2}{\left(\mathcal{T}_1 + \mathcal{T}_2\right)^2} \qquad \text{and} \qquad \mathcal{T}_{min} \simeq \frac{\mathcal{T}_1 \mathcal{T}_2}{4}.$$

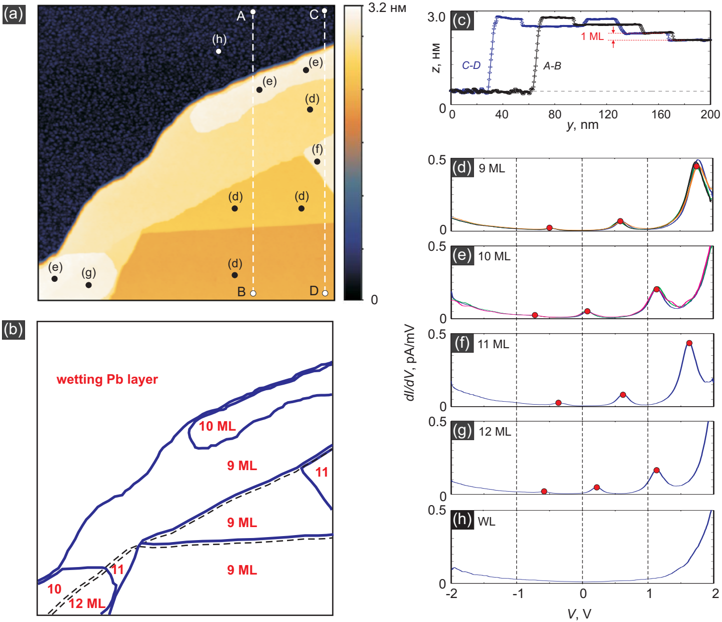
Interference of electronic waves scattered by the first and second barriers:



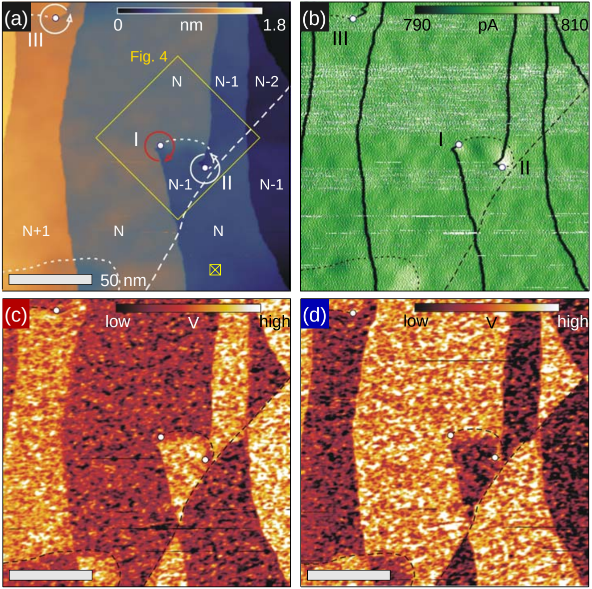
Tunneling via quantum-well states and surface states



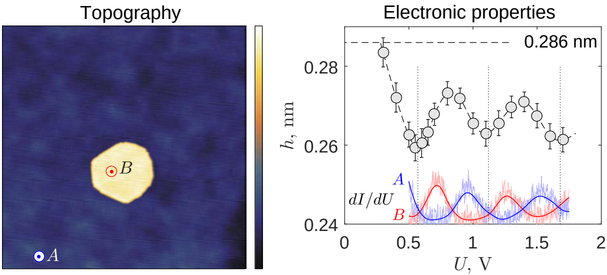
Visualization of subsurface defects in thin Pb(111) films (1)



Visualization of subsurface defects in thin Pb(111) films (2)



Oscillations of visible height of monatomic Pb(111) step



A. Yu. Aladyshkin, Oscillatory bias dependence of the visible height of the monatomic Pb(111) steps: consequence of the quantum-size effect in thin metallic films // Journal of Physical Chemistry C, vol. 127 (27), pp. 13295–13301 (2023)

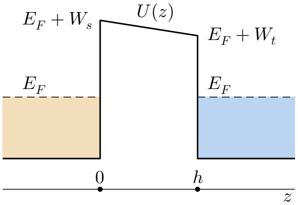
Part IV

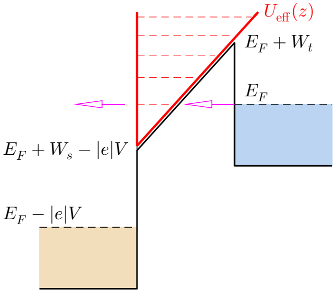
Stark-shifted image-potential states and field-emission resonances

We can neglect the contribution of the image potential and assume that the potential energy near the flat metallic surface has the following linear shape

$$V_{eff}(z) = \begin{cases} \infty & \text{при } z < 0, \\ U^* + F^* \cdot z & \text{при } z \geq 0; \end{cases}$$

with the non-penetrable wall at $z = 0$. Here U is the bias voltage, $U^* = E_F + W_s - |e|U$ is the energy of the bottom of the triangular potential well, and F^* is the effective electrical field near the surface.





Energy of Stark-shifted image-potential states (2)

Reminder: Bohr-Sommerfeld quantization rule for localized electronic states

$$\frac{1}{\pi\hbar}\int_a^b p(z)\,dz = n+\gamma, \qquad n = 0, 1, \ldots$$

where n is the index of the quantum state, and γ is a numerical factor of the order of unity.

We rewrite the Bohr-Sommerfeld quantization rule for a linearly increasing potential in the form

$$\frac{1}{\pi\hbar}\int_a^b p(z)\,dz = n-1+\gamma, \qquad n = 1, 2, \ldots$$

where $a = 0$ and $b = (E - U^*)/F^*$ are classical turning points, m_0 is mass of electron in vacuum, n is integer index, $\gamma \simeq 3/4$.

After substituting $p(z) = \sqrt{2m_0\,(E_n - V_{eff}(z))}$ and integration, we arrive at the spectrum of the quantum-well states in the triangular potential well

$$E_n = E_F + W_s - |e|U + \left\{\frac{3}{2}\,\frac{\pi\hbar}{\sqrt{2m_0}}\,F_n^*\cdot\left(n-\frac{1}{4}\right)\right\}^{2/3}, \qquad n = 0, 1, \ldots$$

Field-emission resonances thin Pb(111) films (1)

Spectrum of quantum-well states in the triangular potential well

$$E_n = E_F + W_s - |e|U + \left\{ \frac{3}{2} \frac{\pi\hbar}{\sqrt{2m}} F_n^* \cdot \left(n - \frac{1}{4} \right) \right\}^{2/3}, \quad n = 1, 2, \ldots$$

where V is the bias voltage.

The process of resonant tunneling via the $n-$th surface electronic state is activated provided that $E_n \simeq E_F$, therefore

$$|e|U_n \simeq W_s + \left(\frac{3}{2} \frac{\pi\hbar}{\sqrt{2m}} \right)^{2/3} \cdot F_n^{*\,2/3} \cdot \left(n - \frac{1}{4} \right)^{2/3}, \quad n = 1, 2, \ldots.$$

Energy spectrum for the higher field-emission resonances ($n \gg 1$)

$$|e|U_n \simeq W_s + \left(\frac{3}{2} \frac{\pi\hbar}{\sqrt{2m}} \right)^{2/3} \cdot F_n^{*\,2/3} \cdot n^{2/3}, \quad n = 1, 2, \ldots$$

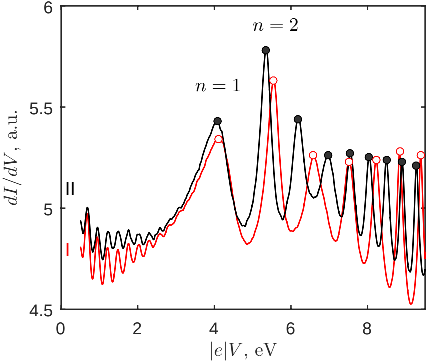
Kolesnychenko, Kolesnichenko, Shklyarevskii, van Kempen, Physica B, v. 291, 246-255 (2000)
Aladyshkin, Journ. Physics: Condens. Matter, v. 32, 435001 (2020)

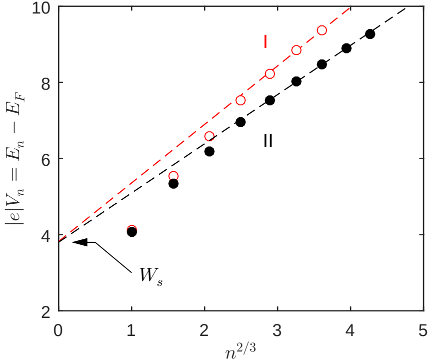
Field-emission resonances thin Pb(111) films (2)

Energy spectrum for the higher field-emission resonances (V_n is the tunneling voltage, $n \gg 1$)

$$|e|V_n \simeq W_s + \left(\frac{3}{2}\frac{\pi\hbar}{\sqrt{2m}}\right)^{2/3} \cdot F_n^{*\,2/3} \cdot n^{2/3},$$

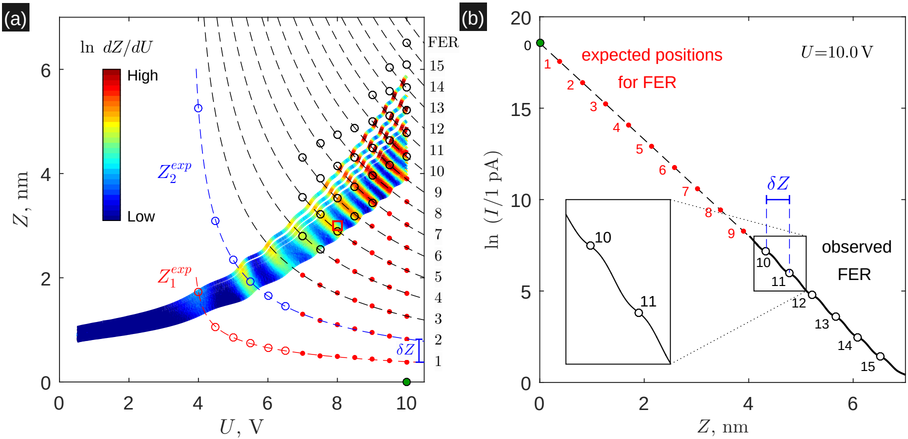
Tunneling spectra for the same Pb(111) terrace, acquired by the STM tip of different shape:





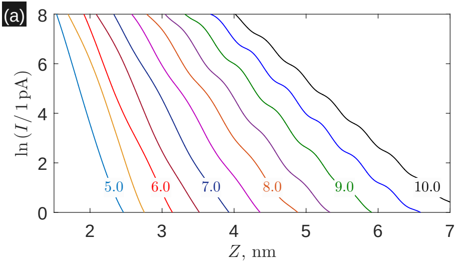
Estimate for local work function $W_s = 3.8 \pm 0.1$ eV

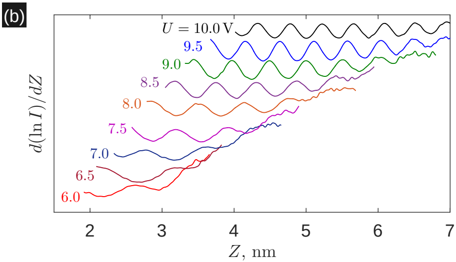
Field-emission resonances thin Pb(111) films (3)



Aladyshkin and Schouteden, *Field-emission resonances in thin metallic films: Nonexponential decrease of the tunneling current as a function of the sample-to-tip distance* // Journal of Physical Chemistry C, vol. 126 (31), pp. 13341–13348 (2022).

Field-emission resonances thin Pb(111) films (4)





Aladyshkin and Schouteden, *Field-emission resonances in thin metallic films: Nonexponential decrease of the tunneling current as a function of the sample-to-tip distance* // Journal of Physical Chemistry C, vol. 126 (31), pp. 13341–13348 (2022).

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