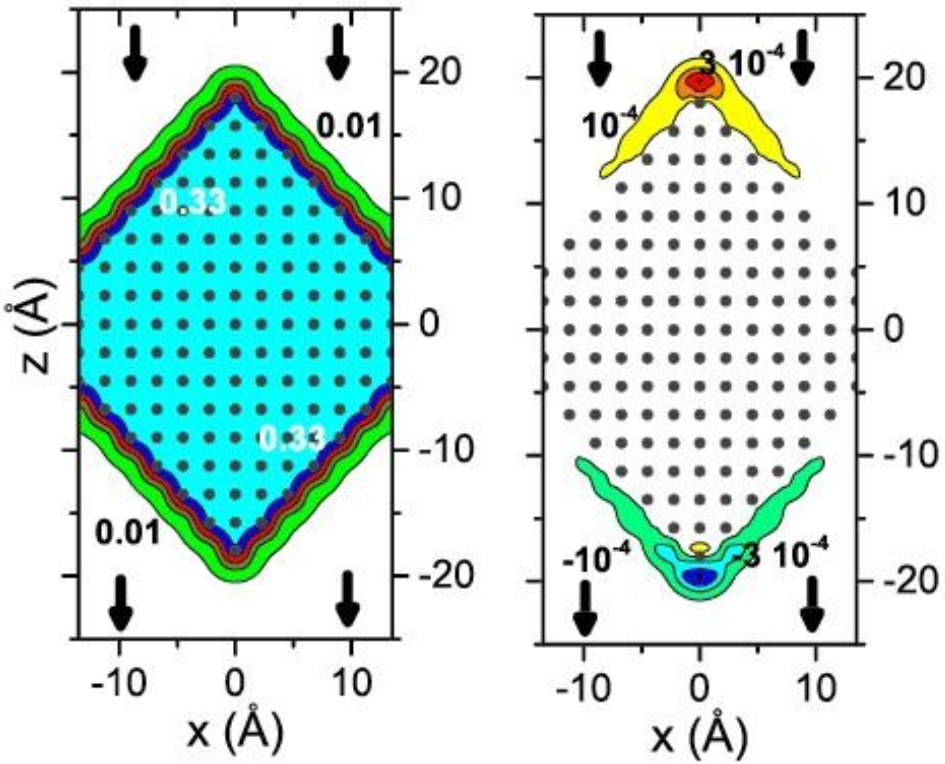
Aims of talk

In vacuum breakdown, there seems renewed interest in the details of how electrostatic fields and potentials vary near charged surfaces.

Modern computing techniques, in particular density functional theory (DFT), but also more classical formulations, are now making very useful progress, —particularly with non-planar surfaces.

Recent Density Function Theory work



J. Lepetit et al, J. Appl. Phys. 120, 085105 (2016).
These authors were aiming to calculate field enhancement factors above "sawtooth" type surfaces, using DFT techniques.

Aims of talk

However, similay scientific problems have arisen before, about 50 years ago, in the context of field ion microscopy.

At that time (and until very recently) we could only deal adequately with planar surfaces, but some useful results were obtained.

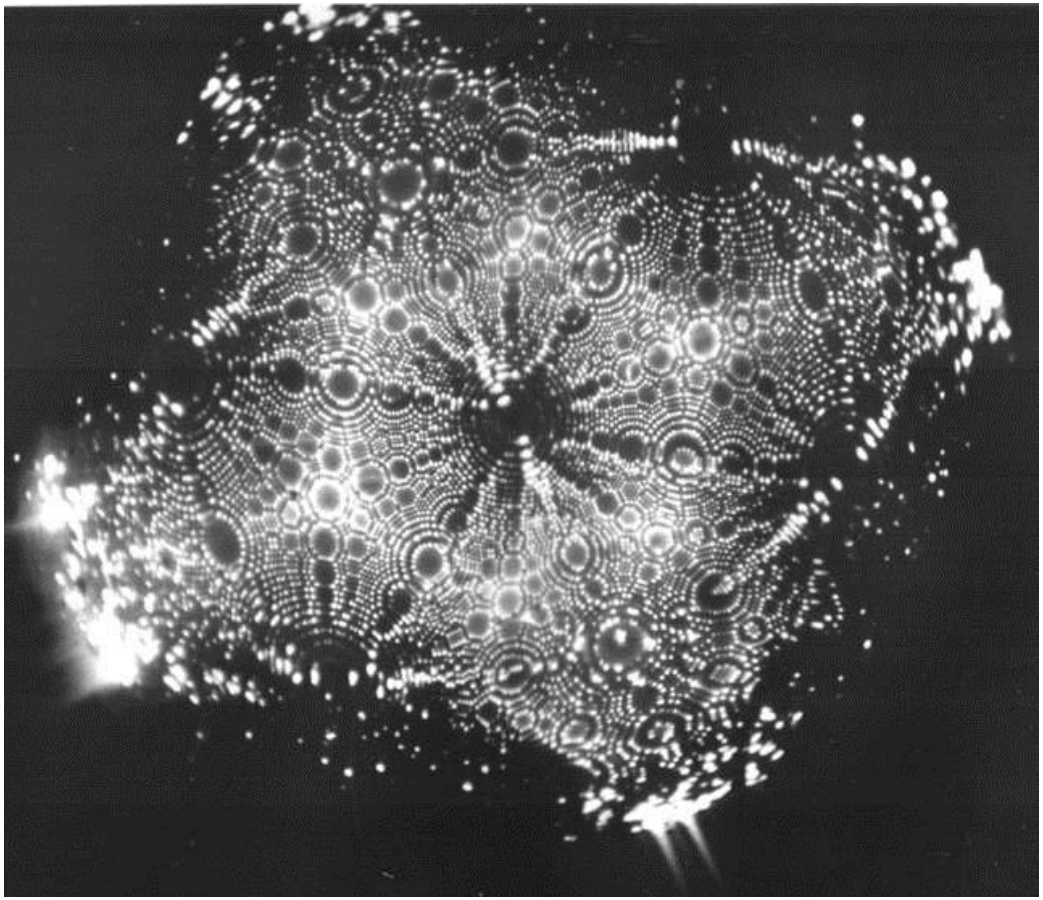
I thought it might be of interest to attempt to relate/compare newer and older theoretical approaches. And to point out that some of the newer techniques could usefully be applied to some of the old problems.

I also consider a question not often asked, namely: exactly where is the tunnelling barrier in field electron emission ?

Structure of talk

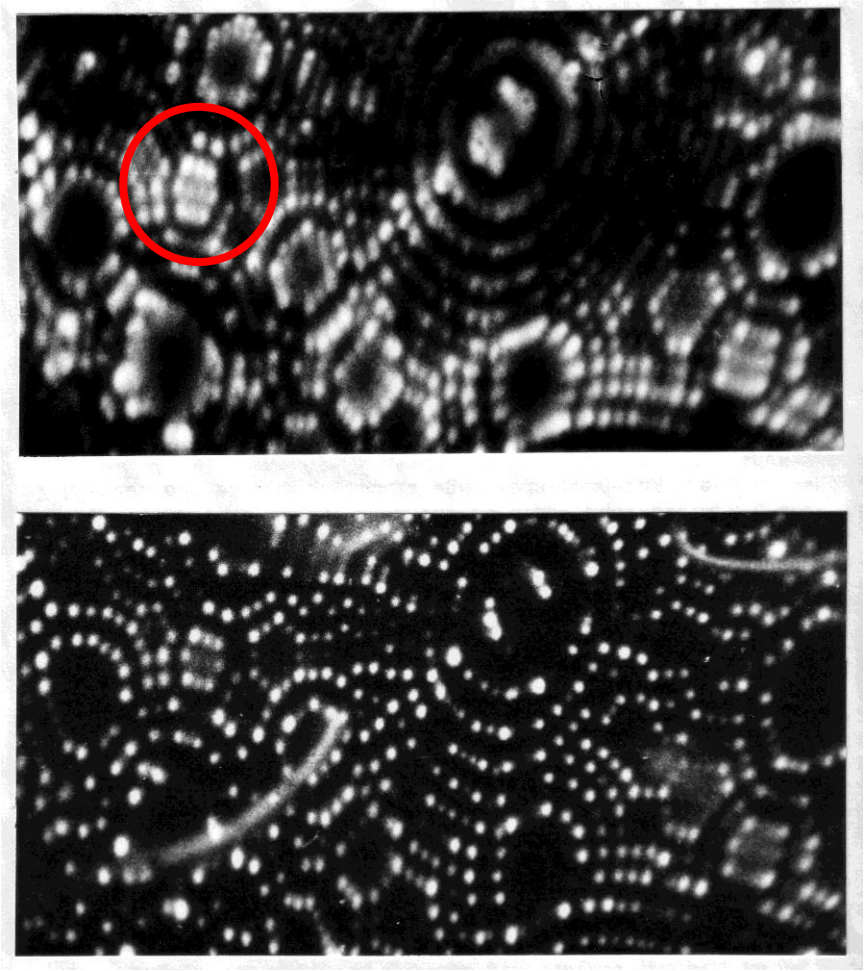
1. Introductory issues
2. The electrical surface and related concepts
3. Numerics and comparisons with recent DFT work
4. Relation to emission phenomena

Field ion micrograph of tungsten point



FIM image: Each white spot is a surface atom. The pattern in the image is related to the arrangement of atoms in the tungsten crystal.

FIM atomic resolution



Near 80 K

Near 5 K

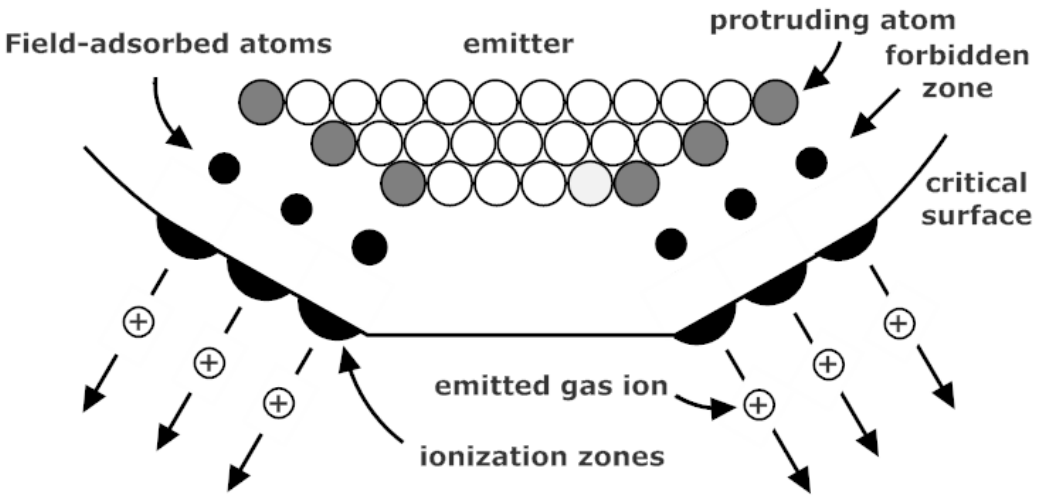
Adjacent atoms in the (111) plane can be resolved.

What were the questions for FIM theory ?

Questions for field-ion-microscope (FIM) theory included:

- **How does the microscope work ?**
- **How does it resolve atoms ?**
- **Where does ionization take place ?**

What were the questions for FIM theory ?

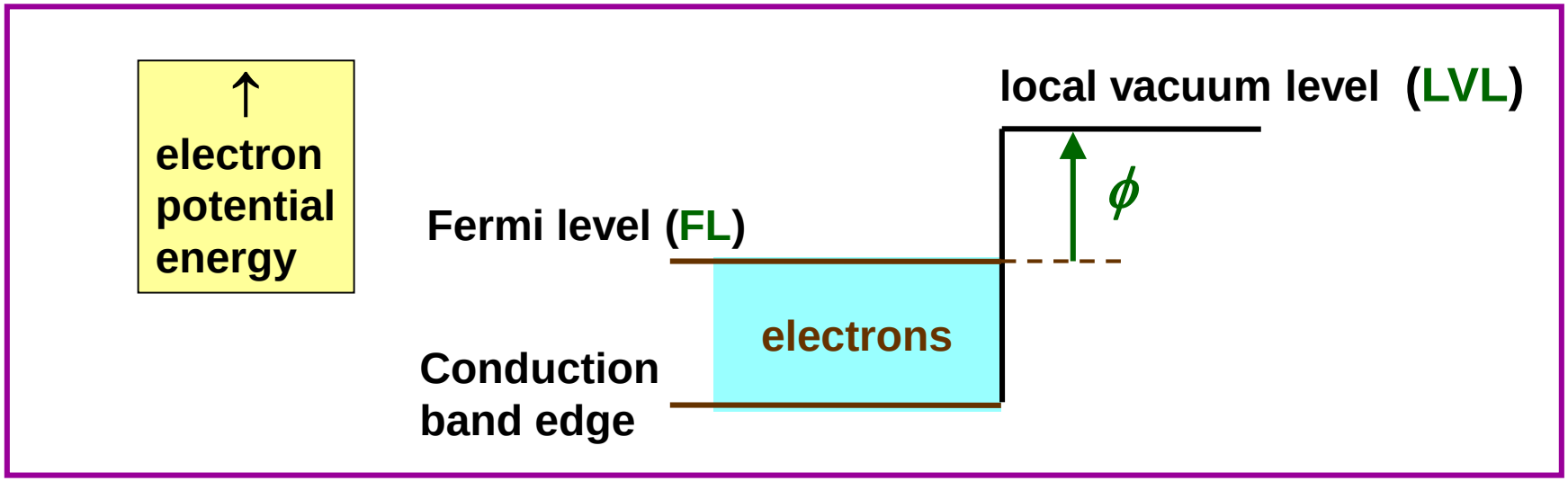


Questions for field-ion-microscope (FIM) theory included:

- **How does the microscope work ?**
- **How does it resolve atoms ?**
- **Where does ionization take place ?**
- **What is the mechanism of field adsorption ?**
- **How does field evaporation take place ?**

The electrical surface and related concepts

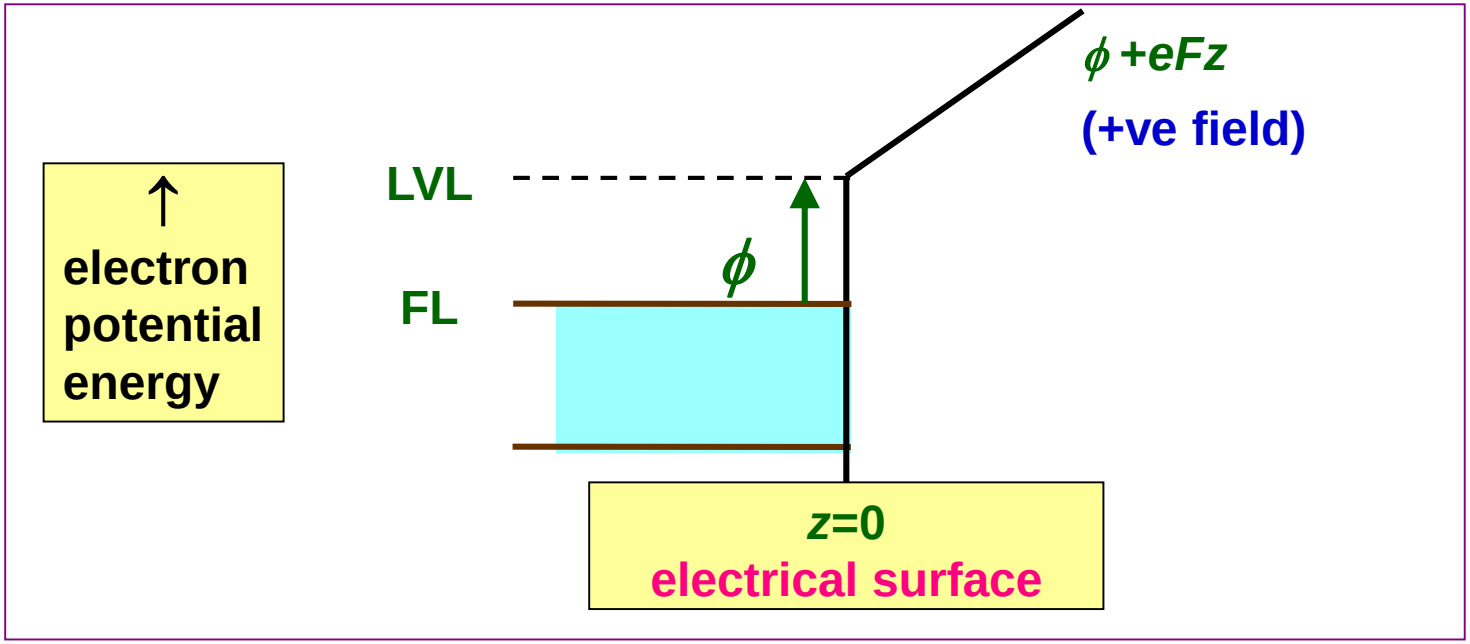
The Sommerfeld model (zero applied field)



A matter of interest is how the electrostatic component of the total electron potential energy (EEPE) varies with position outside a field electron or field ion emitter surface. The argument is initially presented here for a positive field, but the negative-field case is similar.

The simple argument starts from the Sommerfeld model, as shown above, where ϕ is the local work function.

The Sommerfeld model (with applied field)

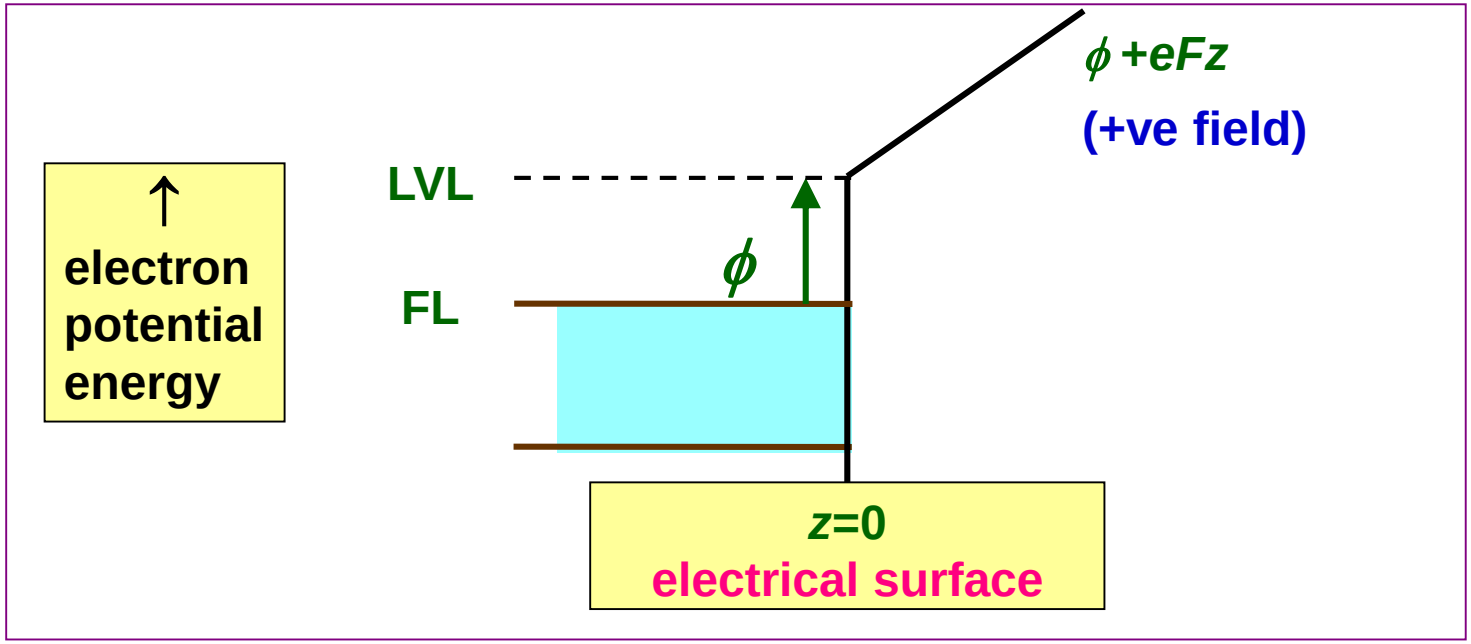


In the limit of large distances *z* from the emitter, and relative to the Fermi level, the *EEPE* is constructed to have the form

$$EEPE = \phi + eFz$$

where: *e* is the elementary positive charge;
z is distance measured from the electrical surface;
***F* is the local electrostatic field (here positive).**

Definitions of the electrical surface - 1



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Definitions of the electrical surface - 2

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In a classical conductor model (as often used, for example, in simulations based on numerical solution of Laplace's equation outside the classical conductor), the electrical surface is identical with the classical-conductor surface.

And the "induced" charge associated with the presence of the external field is regarded as located in an infinitely thin layer at the classical conductor surface (i.e., in the electrical surface).

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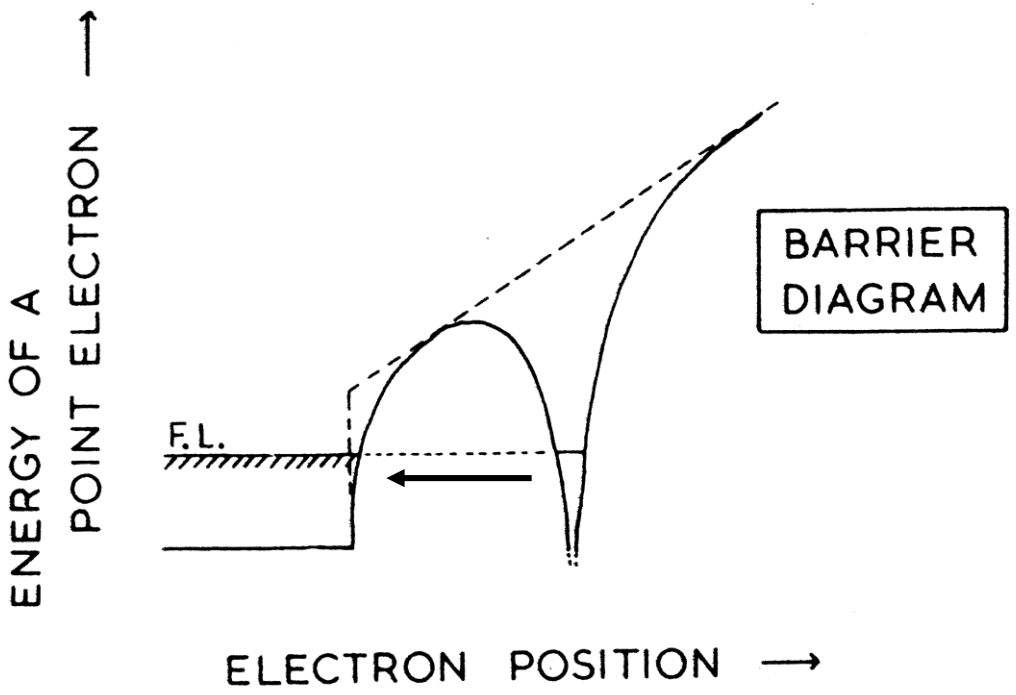
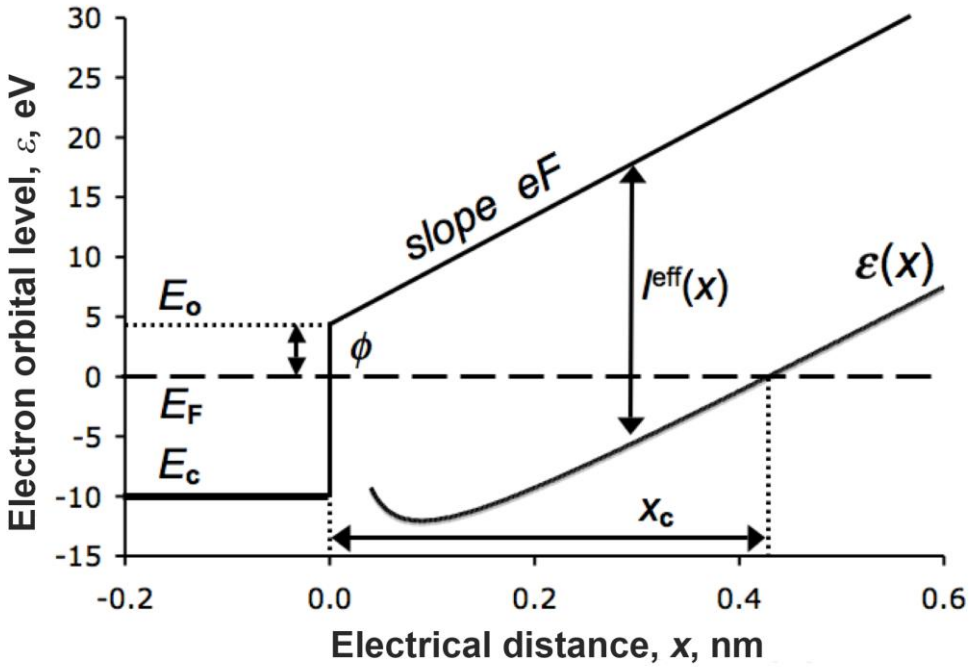
However, an important issue (for both field ion and field electron emission) is "Where is the electrical surface relative to the surface atoms?", because this is part of the answer to the related question of "Where is the tunnelling/transmission barrier relative to the surface atoms ?"

The concept of the critical distance



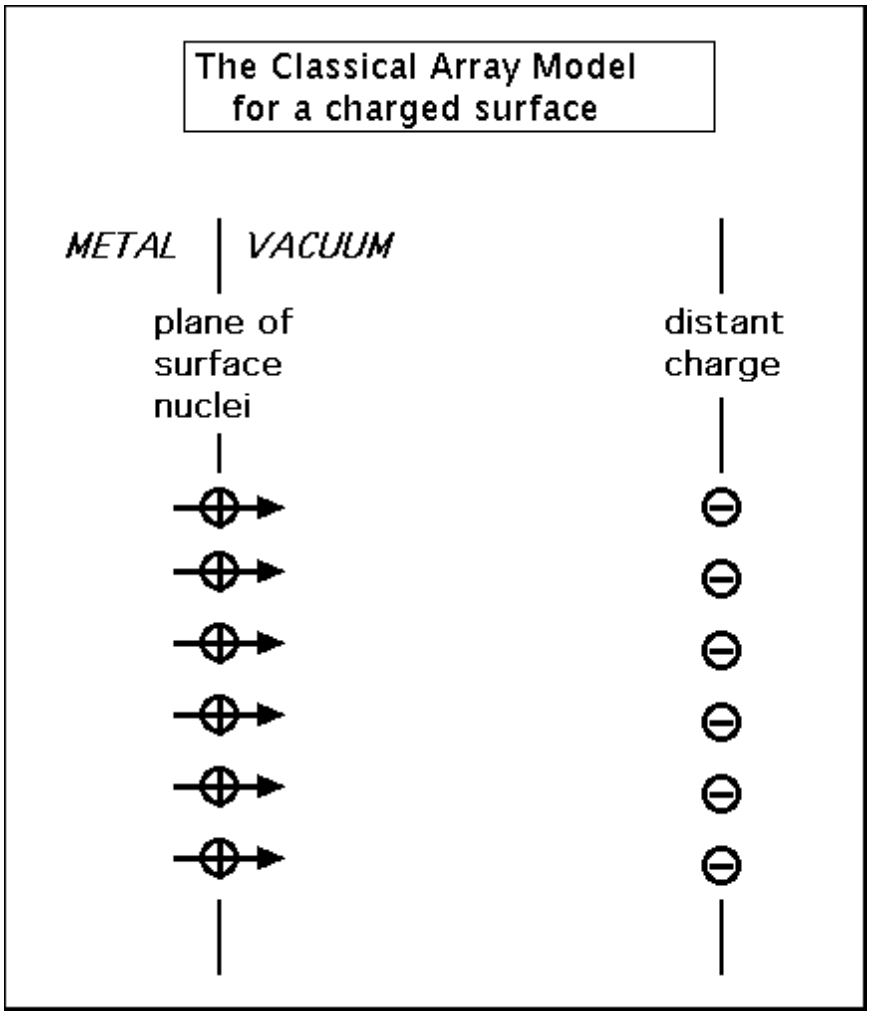
Surface field ionization takes place most rapidly when the topmost electron orbital level in the external gas atom aligns with the Fermi level.

The concept of the critical distance



The critical distance is the smallest distance the electrical surface at which field ionization can occur.

Surface atom polarization as a universal property

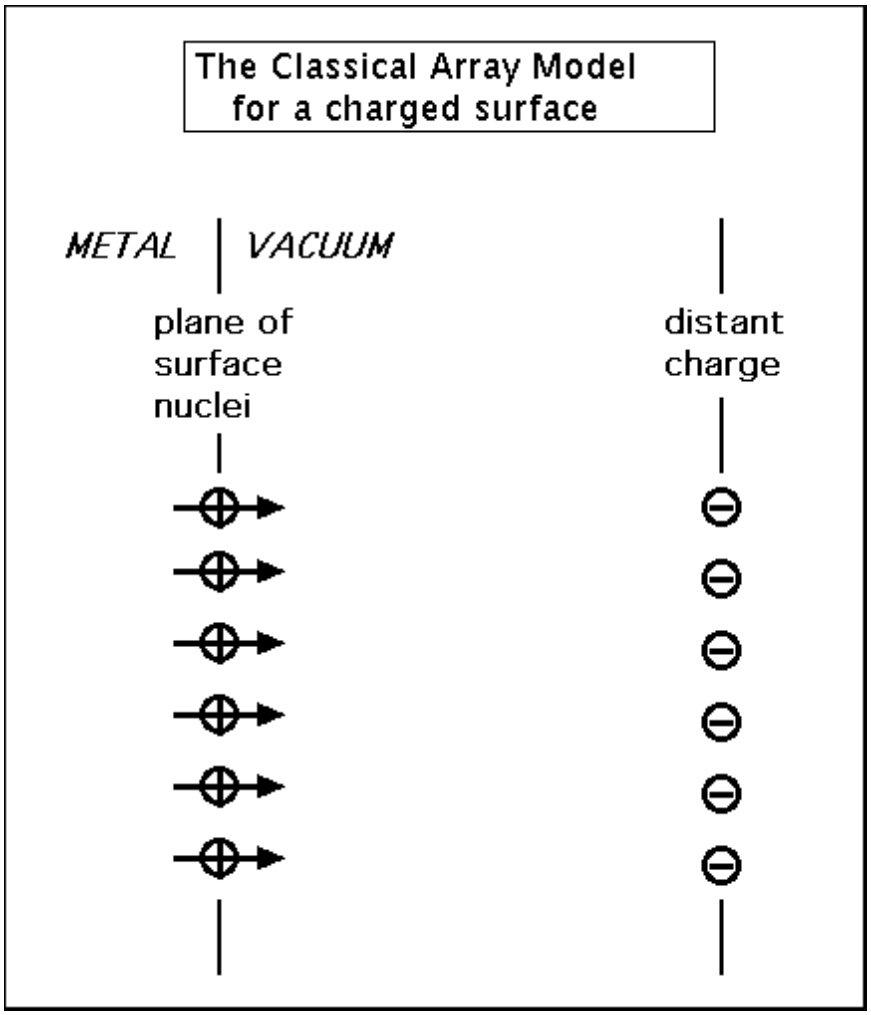


In the planar classical array model, an atomically flat charged surface is modeled by an array of charges & dipoles placed at the positions of the atomic nuclei.

A distant array of charge of the opposite sign represents the counter-electrode [which is needed for electrostatic self-consistency].

The diagram shows a positively charged surface, but the argument is similar for a negatively charged surface.

The planar classical array model



The positive nuclei in the surface atoms are attracted by the distant negative charge.

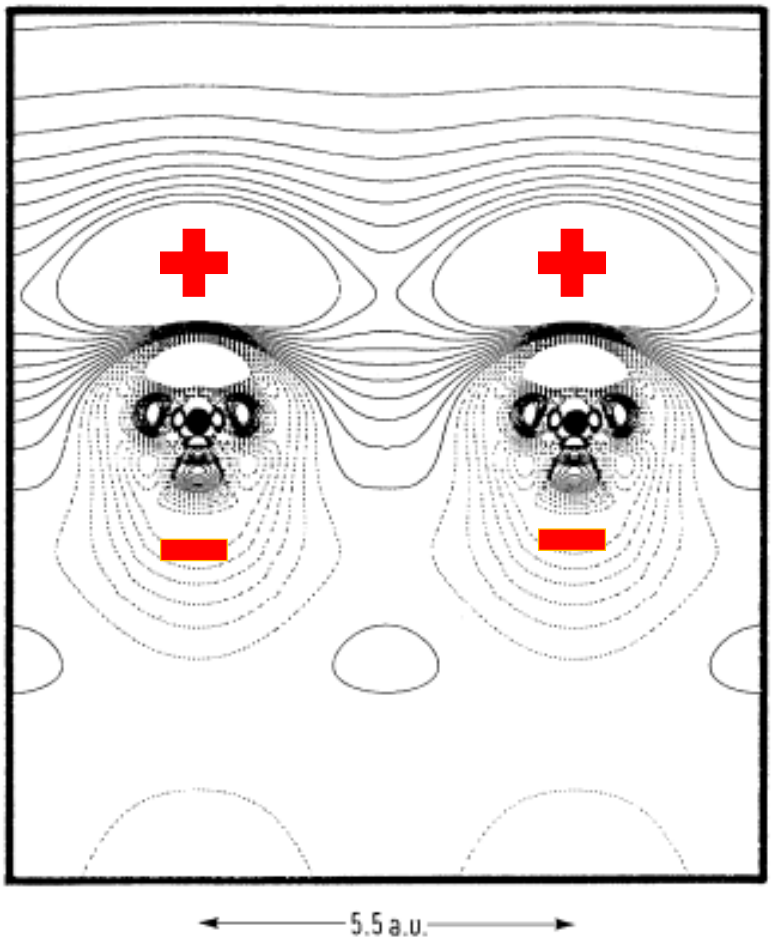
Each surface nucleus moves away from the electrical centre of the electrons in its atom.

This separation causes a restraining pull (by the electrons) on the surface nucleus, towards the emitter.

The equilibrium situation is an electrically polarized atom.

This surface-atom polarization effect is an universal property of charged surfaces.

QM calculations of surface-atom polarization

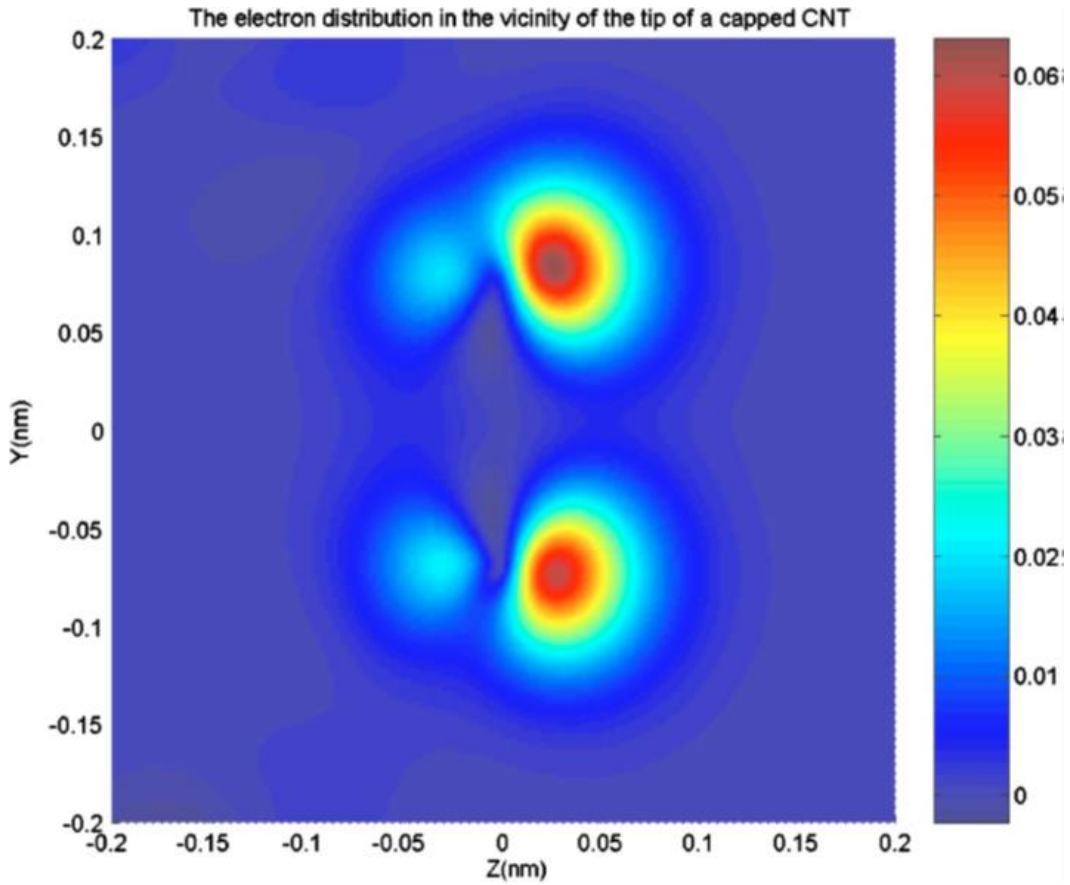


Induced-charge distribution for Ag(001) facet, for an positive external field of 5 V/nm, as calculated by Aers and Inglesfield.

The basic idea that surface atoms might be polarized was introduced (in the context of FEM adsorption studies) in the 1950s.

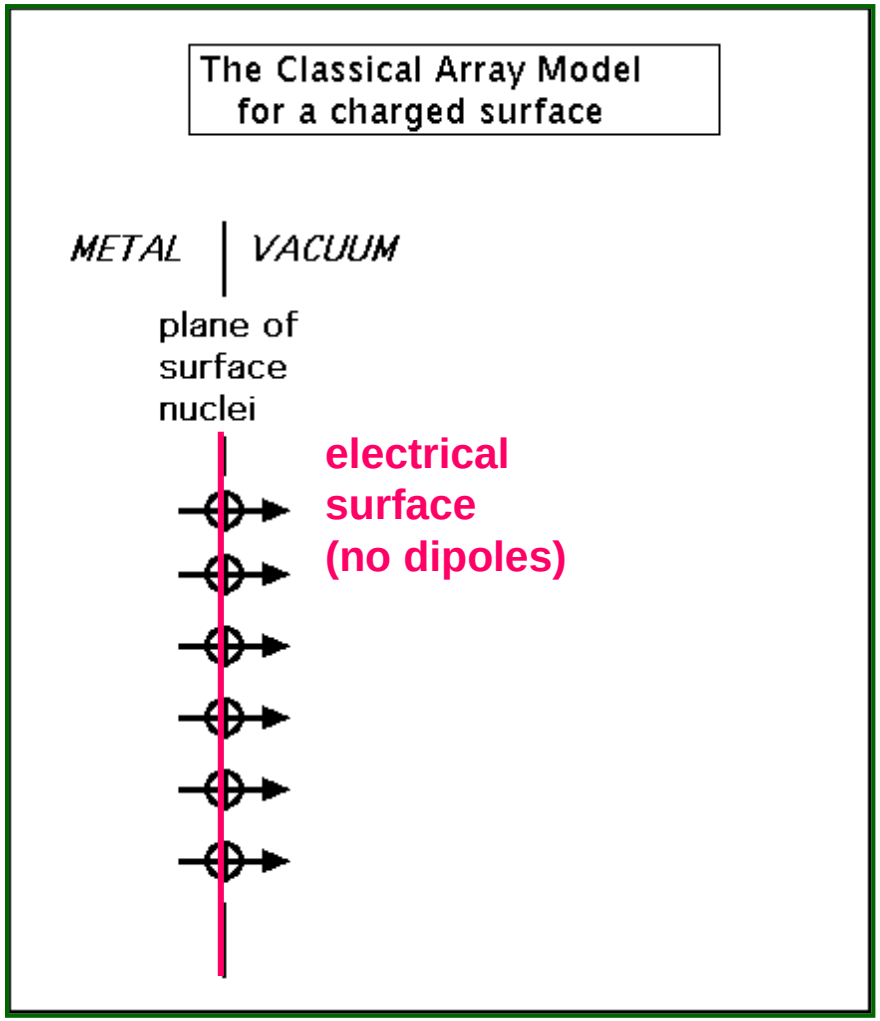
But it took until the late 1980s before the first satisfactory quantum-mechanical (QM) calculations of the effect (shown alongside) were done.

QM calculations of surface-atom polarization



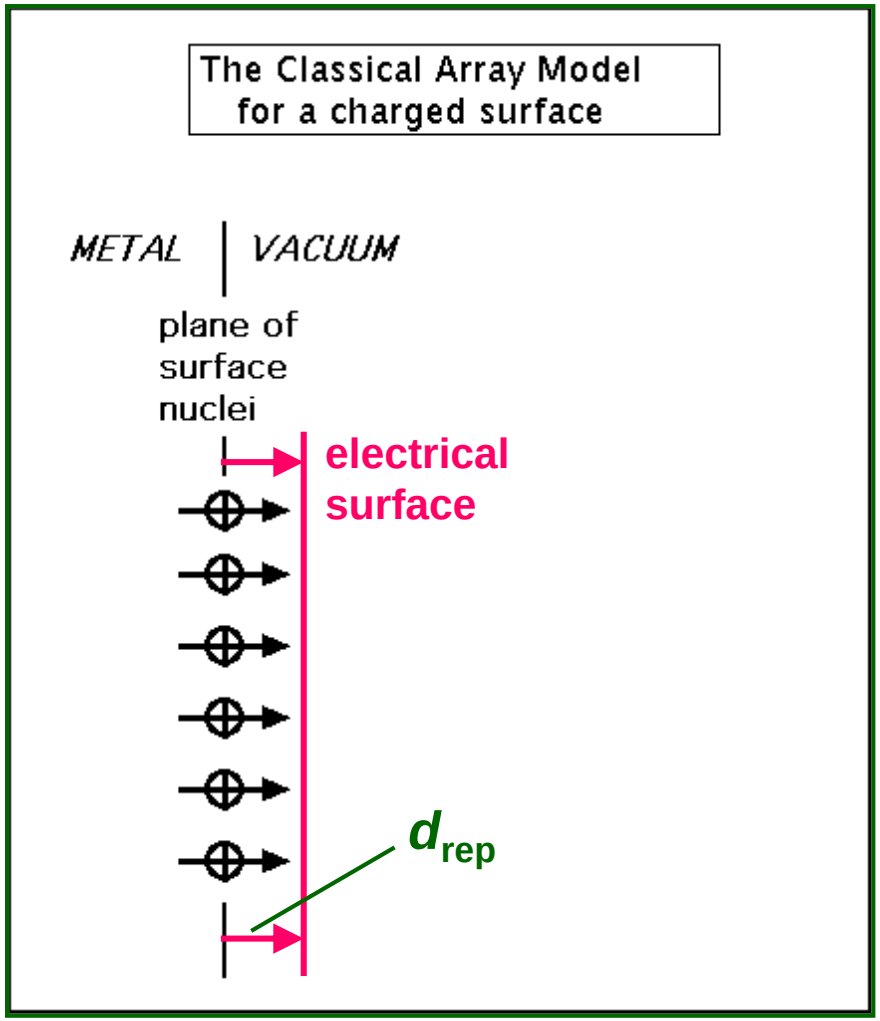
Surface-atom polarization effects of this kind also occur with negative applied fields, as demonstrated by J. Peng et al. in first principles calculations on closed carbon nanotubes.

The concept of repulsion distance



It can be shown that, in the absence of surface dipoles, the z=0 plane would lie in the plane of the surface nuclei.

The concept of repulsion distance

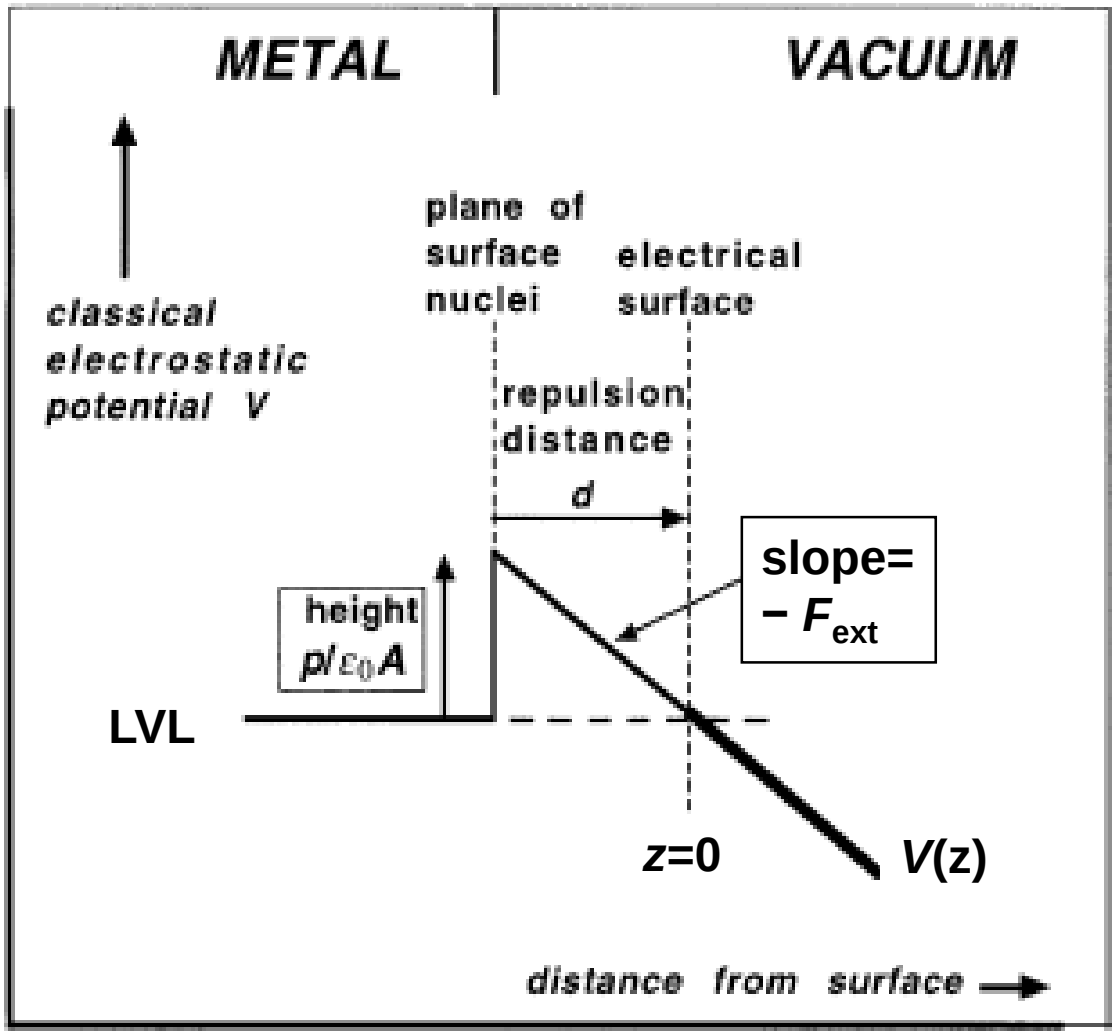


It can be shown that, in the absence of surface dipoles, the z=0 plane would lie in the plane of the surface nuclei.

The presence of the dipoles moves the electrical surface outwards (towards the vacuum) by the repulsion distance d_{rep}.

The size of d_{rep} is comparable with atomic radii, as assessed by half the nearest-neighbour distance.

The concept of repulsion distance



Classical formula for repulsion distance

An analysis of the electrostatics enables a formula to be obtained for d_{rep} , in terms of the surface crystallography and effective surface-atom polarizability α_{pol}. For cubic-system crystals this takes the form

$$d_{rep} = p/\varepsilon_0 A_s = \tfrac{1}{2}\alpha_{pol}/[\varepsilon_0 A_s(1+T_{str}\alpha_{pol}/4\pi\varepsilon_0 C_{lat}^3)]$$

where C_{lat} is the bulk lattice parameter, A_s is the surface area per atom, and T_{str} is a structure factor that depends on the crystallographic structure of the relevant crystal face.

In the case of Al, these classical results can be compared with the QM calculations of Aers and Inglesfield, as shown on next slide

Comparison of QM and classical models

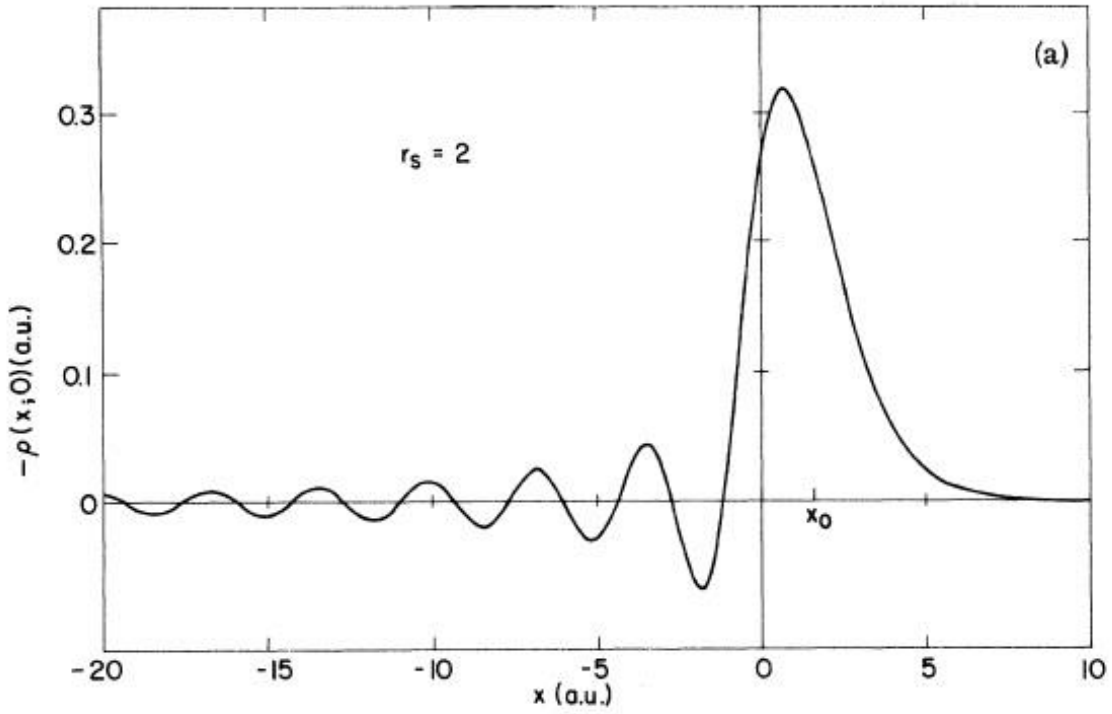
Table 1
Values for the repulsion distance d, calculated for different faces of aluminium by means of self-consistent quantum mechanical (q/m) calculations and the classical array model

Reference	Material & facet	Repulsion distance (pm) at zero field	
		q/m model	Classical model
Lam and Needs [10]	Al (1 1 1)	167	150
Inglesfield [6]	Al (1 0 0)	159	152
Lam and Needs [10]	Al (1 1 0)	152	149

In all cases, classical and quantum-mechanical values agree to within 20 pm.

This provides justification for using the classical model to estimate values for other metal systems, as shown shortly.

QM calculations of surface-atom polarization



An equivalent result had been obtained earlier by Lang and Kohn, using a so-called "jellium" model.

This showed that the electrical surface was OUTSIDE the jellium edge.

Definitions of the electrical surface - 3

In their one dimensional jellium model, Lang and Kohn showed that the electrical surface in the centroid of the induced charge.

I have shown that this is also true for the planar array model.

Numerics and

Comparisons with recent DFT work

Repulsion distances for some metal systems

Table 1
Values for the repulsion distance d, for the close-packed faces of selected metals. Also shown are the volume polarisability α_v as tabulated in Ref. [17], and the atomic radius as estimated by half the nearest-neighbour distance R_{NN} in the solid metal

Material and face		$R_{NN}/2$ (pm)	α_v 10^{-30} m^3	d (pm)
W	bcc (1 1 0)	137	11.1	157
Ir	fcc (1 1 1)	136	7.6	144
Mo	bcc (1 1 0)	136	12.8	160
Pt	fcc (1 1 1)	139	6.5	141
Au	fcc (1 1 1)	144	5.8	138
Rh	fcc (1 1 1)	135	8.6	147
Fe(α)	bcc (1 1 0)	129	8.4	143
Ni	fcc (1 1 1)	125	6.8	136
Cu	fcc (1 1 1)	128	6.1	135

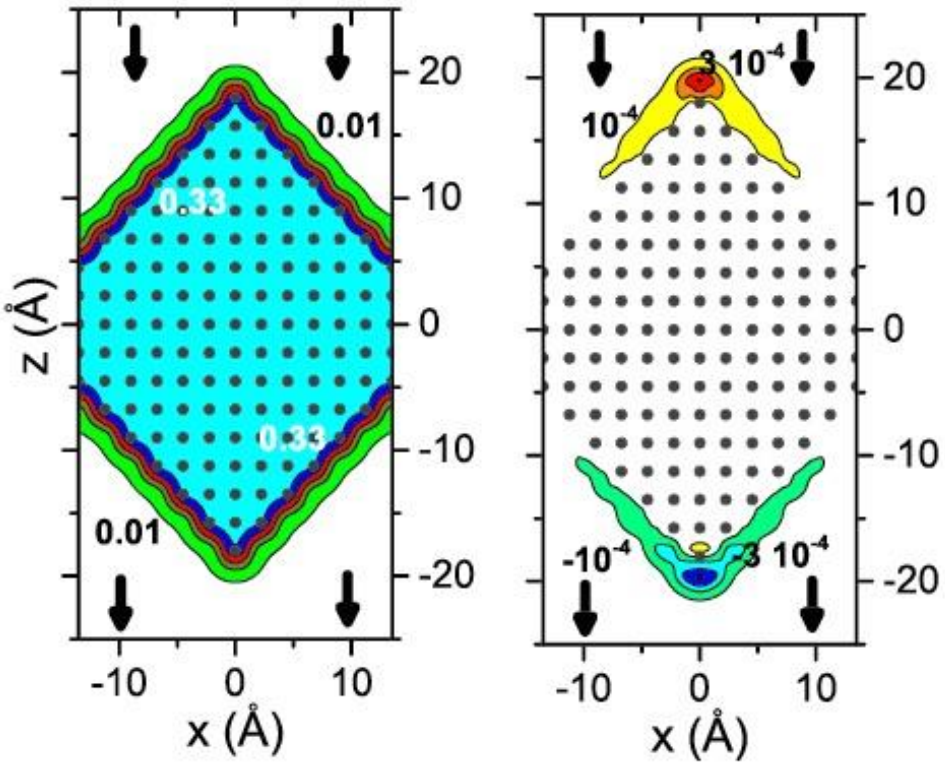
Comments on repulsion-distance calculations

In general, repulsion distances are comparable in size (often slightly greater than) atomic radii.

For tungsten, the predicted electrical surface position tends to be confirmed (at least qualitatively) by experimental measurements of field ion appearance energy.

Comparison with recent DFT work

085105-4 Lepetit, Lemoine, and Márquez-Mijares



The "induced charge barycentre" of Lepetit et al. is essentially the same concept as the older concept of the electrical surface, when this is interpreted as the centroid of the induced charge.

Comparison with modern DFT calculations

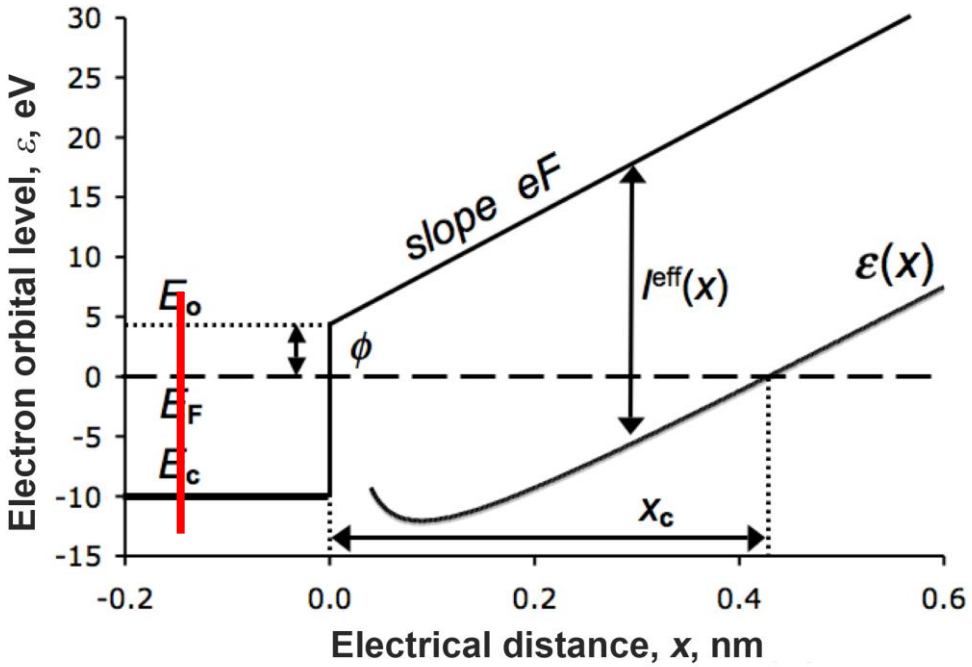
Theoretical estimates of repulsion distance for tungsten are:

Classical array model – (110) surface:	**157 pm**
DFT (sawtooth apex – positive field):	**161 pm**
DFT (sawtooth apex – negative field):	**213 pm**

Good general consistency between all models, QM and classical.

Relation to emission phenomena

Surface field ionization



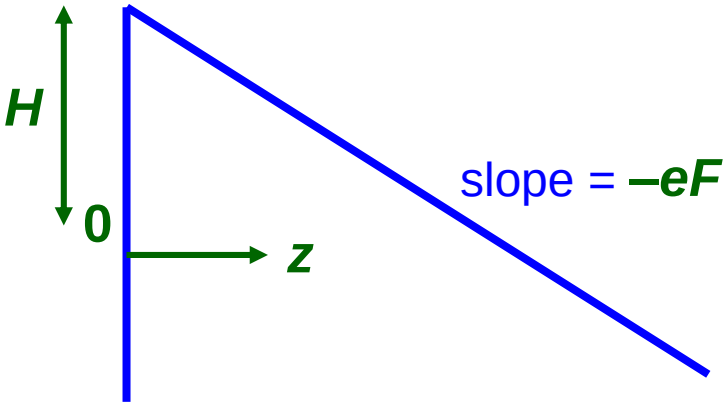
For helium on tungsten, work function = 4.5 eV, ionization energy = 24.6 eV, best image field = 45 v/nm, so critical distance is about 450 pm.

Repulsion distance is about 160 nm.

Distance of ionization zone from surface-atom nuclei is about 600 pm.

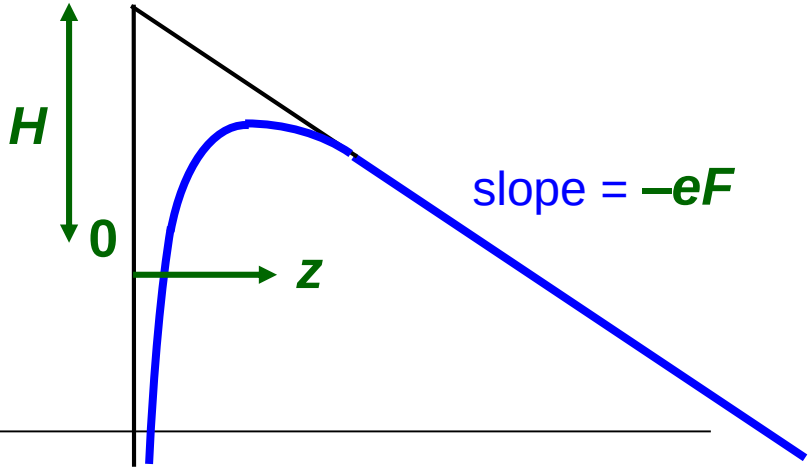
Two special barrier forms

Exactly triangular (ET) barrier



$M(z) = H - eFz$

Schottky-Nordheim (SN) barrier



$M(z) = H - eFz - 1/16\pi\varepsilon_0 z$

Repulsion distance is about 200 nm (according to Lepetit et al.)

For SN barrier, distance from electrical surface to inner edge of barrier is about 150 pm, under typical emission conditions.

Corresponding distance from surface-atom nuclei is roughly 350 nm

Barrier appears to be significantly outside atomic wave-functions.

Thanks for your attention