RESOLUTION OF FIELD-ION MICROSCOPY VERSUS SCANNING TUNNELLING MICROSCOPY FOR OBTAINING SURFACE CHARGE DENSITY CORRUGATIONS

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RESOLUTION OF FIELD-ION MICROSCOPY VERSUS SCANNING TUNNELLING MICROSCOPY FOR OBTAINING SURFACE CHARGE DENSITY CORRUGATIONS

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Résumé - La densité de charge au-dessus d'une surface métallique ondulée est calculée afin de comparer la résolution du field-ion microscope (FIM) et le scanning tunnelling microscope. La surface est représentée par un modèle jellium. Les calculs indiquent que le FIM pourrait donner la meilleure résolution. Il est suggéré que le FIM peut être utilisé pour déterminer l'ondulation de la densité de charge au-dessus de la surface et donc pourrait élucider la structure de la cellule unitaire de la surface.

Abstract - Our investigations of the resolution of the field-ion microscope (FIM) and the scanning tunnelling microscope, based on calculations of charge density above a structured metal surface represented by a jellium model, indicate that better resolution may be expected in the FIM. We suggest that FIM can be used to determine the corrugation of the charge density above a surface and hence to elucidate the structure of a surface unit cell.

For many years Field-Ion Microscopy (FIM) /1/ had the unique distinction of being the only known means of directly viewing the structure of solid surfaces in atomic detail. This distinction has now been challenged by Scanning Tunnelling Microscopy (STM) developed by Binnig, Röhrer, Gerber and Weibel /2/. STM has attracted much attention recently because of its versatility and remarkable resolution of surface topography. FIM may not be able to compete with the versatility of STM but, we believe, it can provide similar information on the surface topography, in the cases to which it can be applied, but with potentially better resolution and with a substantial improvement in the reproduciblity of results. Our calculations indicate that FIM can be used as a powerful technique to measure the corrugation of the charge density above a surface in real space.

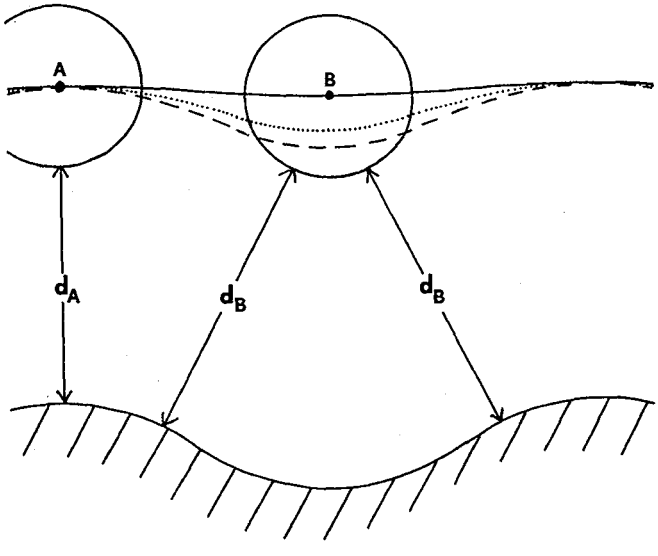
1. IMAGING IN FIM AND STM

Both FIM and STM rely on an electron tunnelling mechanism for their operation, the similarities of the two techniques have been pointed out by Kingham et al. /3/. Neither technique is completely understood theoretically, but we have been careful to use a similar theoretical approach in each case in order to compare fairly the two techniques. In FIM it is known that protruding surface atoms, usually at the edge of a net plane, are imaged and in some cases all the atoms in a small net plane can be imaged. Various explanations have been proposed for this imaging which occurs by ionization of gas atoms at or close to the critical distance for field ionization. The simplest explanation, which has recently been shown to be consistent with experimental observations /4/, is that the critical equipotential surface is closest to the metal surface where the surface protrudes, and that the tunnelling barrier is thus shortest at that point. Alternatively, but equivalently, there is a bulge in the surface charge density above a protrusion so that there is greater overlap between the wave functions of a gas atom at the critical distance and the metal wave functions. This alternative view demonstrates the similarity with scanning tunnelling microscopy rather better. In STM the probe scans over the surface with its height continuously adjusted to give a constant tunnelling current (i.e. at constant wave

function overlap). In fig. 1 we show lines of equal charge density compared to the equipotentials which are much less corrugated, and to the line scanned by the tip of the STM at constant tunnelling current.

Imaging in FIM depends on the difference in ionization rates at the critical distance above the protrusions compared to the mid-point between protrusions (pts A and B in fig. 1), whereas imaging in the STM is by monitoring the height of the tip as it scans along the lines of constant tunnelling current. If the STM tip was a point, rather than a few atoms across, it would scan along the lines of constant current density according to the transfer Hamiltonian model of Tersoff and Hamann /5/ and also according to the scattering model of Garcia et al. /6/. However, because of the finite (and unknown!) size of the tip it does not measure the full corrugation of the charge density. In the FIM case the tunnelling probe is a single gas atom and so the resolution is improved due to the small probe size and the reproducibility is improved because the probe is well characterised.

Conventionally ions in a field-ion microscope are detected visually by their impact on a phosphur screen, but it is quite feasible to detect the ions using counters with single ion sensitivity. By careful positioning of a probe hole with reference to a field-ion image the high magnification available using FIM means that it is quite possible to count the number of ions originating from a region of the surface of sub-ångström dimensions. It is thus a real possibility, well within present experimental capabilities, to measure the number of ions originating from different regions of the surface unit cell. In a standard field-ion microscope operating at Best Image Voltage (BIV) it is usually not possible to see any detail of the centre of a net plane (only the edge atoms are imaged), however, there will still be enough ionization occurring in the middle of at least some crystal planes for detectors with



——— critical equipotential
- - - - - line of constant charge density
..... line scanned by STM tip

Fig. 1 - The critical equipotential, compared to a line of constant current density and the line scanned by an STM tip at constant tunnelling current above a model metal surface. Imaging gas atoms are shown at A and B. We note that at position B there are two directions of maximum tunnelling current.

high resolution and single particle sensitivity to reveal differences in ionization probability. From these differences in ionization probability it is possible to deduce accurately the corrugation of the charge density above the surface and hence to elucidate the structure of the surface unit cell.

We are well aware that resolution in the FIM is ultimately limited by the uncertainty principle, and that there is also a limitation due to a thermal lateral velocity of imaging gas atoms when they are ionized /7/, but even so, particularly with recent advances in image processing, genuine sub-atomic resolution seems possible. There are also potential problems due to gas supply effects and to field adsorbtion, but whereas these problems certainly affect imaging of kink step atoms, they should not be serious in the middle of a plane.

2. MODEL AND CALCULATIONS

Garcia et al. /6/ have recently presented an exactly solvable theory for the tunnel current between two electrodes of general shape described by a jellium model. This theory is applicable to both STM and FIM where one of the electrodes is instead an imaging gas atom.

Details of the model used and the application of this theory to STM are given by Garcia and Flores /8/. In fig. 2 we show the model used here for the calculations relevant to FIM. The real potential along the direction of a surface normal, relative to a zero at the metal Fermi level, is given to a good approximation by

$$V(z) = \emptyset + Fz + 1/(s+z) - 1/4z - 1/(s-z) \qquad (1)$$

in atomic units, including image potentials, but we approximate this by a "top-hat" shape with height $\emptyset_B$. In our model potential we have substituted an abrupt potential

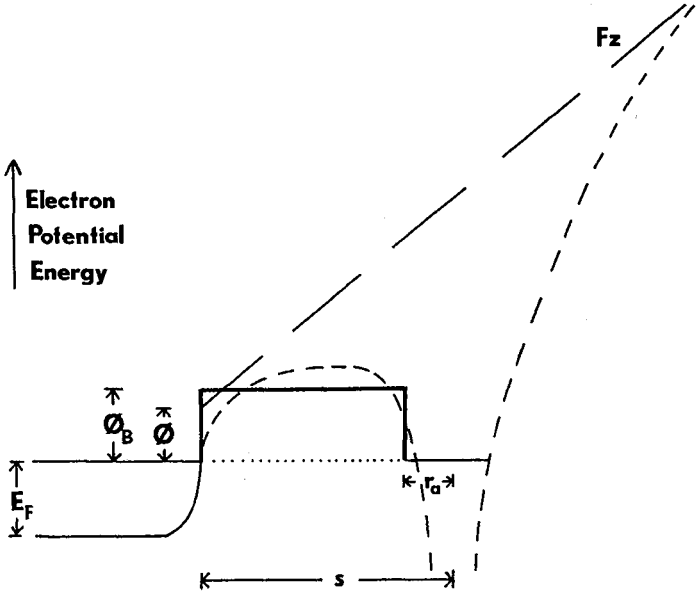


Fig. 2 - The model potential barrier adopted for this work ———— compared to a more realistic potential — — — — . The imaging gas atom (of radius r_a) is shown at the critical distance when its energy level is equal to the Fermi level.

for the real surface potential and we also impose an abrupt potential at the atomic radius as shown in fig. 2. The barrier height, ϕ_B, is found by taking the average value of V(z) from eq. 1. Due to the applied field, ϕ_B is larger than the barrier height in STM.

We now follow an argument given by Garcia and Flores /8/ for STM to derive an expression for the ionization rate constant, I, as

$$I = C\, R_{eff} \exp[-2.14kd] \qquad (2)$$

where C is a constant depending on the metal density of states at the Fermi level, $k = (2m\phi_B/\hbar^2)^{1/2}$ and d is the minimum barrier distance. Garcia and Flores carefully verify the validity of this approximate formula by comparison with their exact calculations. The value 2.14 rather than 2.0 appears in the exponential because Fermi level electron wavefunctions with wave vector not parallel to the surface normal are included and these decay faster into the barrier. R_{eff} in eq. 2 is the effective radius of curvature of the atom-surface system defined by

$$1/R_{eff}^{\ 2} = (\ 1/r_a + 1/R_1^{\ s}\ )\ (\ 1/r_a + 1/R_2^{\ s}\ ) \qquad (3)$$

where r_a is the atomic radius and $R_1^{\ s}$ and $R_2^{\ s}$ are the principle radii of curvature of the surface at the minimum distance between atom and surface. Using eq. 2 it is now a fairly simple matter to calculate the relative ionization rates at the critical distance, above surface protrusions compared to the mid-point between protrusions, except that we have not yet determined the locus of the critical surface. In fact the critical surface is much less corrugated than the lines of equal charge density at a similar distance from the surface /4/, indeed it is the difference in the corrugation of equipotentials and equi-charge-density-lines that gives the contrast in FIM. Homeier and Kingham's model calculations /4/ can be adapted to give an approximate expression for the corrugation amplitude, a_c, of the critical surface which is

$$a_c = a_s^{\ 2} / z_c \qquad (4)$$

for a surface corrugated in one direction only where a_s is the corrugation amplitude and z_c is the mean critical distance, or

$$a_c = a_s^{\ 3} / z_c^{\ 2} \qquad (5)$$

for a surface corrugated equally in both directions. As a_s is typically $\ll z_c$ the corrugation of the critical surface is small (and is almost negligible in the case when we consider whether a small surface corrugation could actually be measured).

Our first calculation is for a Au (110) 2x1 surface which is reconstructed by a missing row model /9/ and which we represent by a sinusoid of amplitude 1.62 Å and wavelength 8.14 Å in one direction. We ignore any corrugation in the other direction (along the rows). The second calculation is for a much less corrugated surface represented by two sinusoids (one in each direction) of amplitude 0.2 Å and wavelength 4 Å, which is reasonably typical of (100) faces of fcc or bcc crystals, (Godby R., private communication).

3. RESULTS AND DISCUSSION

Results for the Au (110) 2x1 surface are given in part (i) of table 1 including comparable results for STM assuming that the STM tip is scanned along the critical surface, rather than at constant tunnelling current. The results show that remarkably good contrast should be obtainable using He with the contrast being reduced, but still good, moving down the list of imaging gases. This trend is caused by two factors:

i) Contrast is improved by a high work function associated with a high electric field.

ii) The smaller the surface probe the better the contrast.

We note that the best image field for He may be too high for (e.g.) an Au tip to be stable, but the results are still shown for completeness and in any case with a technique of ion counting it should be possible to work at fields below the conventional best image field. In practice the actual contrast achievable with Ne may be better than with He because the effects of both uncertainty principle and lateral thermal velocity are less for the more massive Ne.

The results of our second calculation are shown in part (ii) of table 1. Although the contrast is much less, it is still sufficient to allow the surface structure of an atomically smooth, close-packed surface to be seen by counting individual ions, though it may be necessary to count a large number of ions to reduce statistical errors

Table 1

A comparison of the calculated contrast attainable in FIM, for a model representing, (i), the Au (110) 2x1 surface and (ii), a (100) face of an fcc or bcc crystal, using various inert imaging gases. A comparable figure for STM, assuming a 3 Å radius tip, is also given. In case (ii) our calculations indicate no significant contrast for Xe or for an STM tip. I_A and I_B are the ionization rate constants at positions A and B which correspond to barrier widths d_A and d_B.

Imaging Gas	Field Strength (V/Å)	Barrier Height Φ_B (eV)	(i)				(ii)			
			d_A(Å)	d_B(Å)	d_B-d_A(Å)	I_A/I_B	d_A(Å)	d_B(Å)	d_B-d_A(Å)	I_A/I_B
He	4.5	6.6	2.36	3.55	1.19	10.7	2.97	3.17	0.20	1.53
Ne	3.5	5.6	2.48	3.67	1.19	7.9	3.09	3.28	0.19	1.29
Ar	1.9	4.1	3.24	4.32	1.08	3.9	3.85	4.03	0.18	1.08
Kr	1.5	3.6	3.52	4.55	1.03	3.0	4.13	4.31	0.18	1.02
Xe	1.2	3.1	3.38	4.33	0.95	2.3	3.99	4.17	0.18	≅1
STM(a)	---	3.0	4.00	4.85	0.85	2.0	4.00	4.17	0.17	≅1

(a) assuming a 3Å radius tip

4. FURTHER EXPERIMENTAL POSSIBILITIES

A possible criticism of a field-ion microscopy determination of surface structure is that the field may alter that structure. Indeed in the case of W (100) there is strong evidence indicating a field-induced reconstruction from p(1x1) to c(2x2) /10/ which occurs by field evaporation of alternate atoms. If such a change of structure does occur then by using different imaging gases, with a wide range of ionizing field from 4.5 V/Å for He to only about 1.2 V/Å for Xe it should be possible to measure the change in surface corrugation with applied field and hence to deduce something about the shape of the potential well of surface atoms. Counting individual ions offers an extra advantage here, that a range of field strength up to BIF could be used for each imaging gas. We note that it is not feasible to work at fields much above BIF because imaging gas atoms are then ionized before they can accomodate to the tip temperature and the resolution is thus inferior /7/.

There is some uncertainty in field-ion microscopy about why electro-negative adsorbates such as oxygen are not seen. It may be that they are field desorbed or else migrate into the bulk so that they are not present on the surface, or perhaps they are present, but are simply invisible in conventional FIM. If the latter is true then our calculations indicate that ion counting should reveal the positions of adsorbates in FIM. Observations of O on Ni (110) have recently been reported using STM /11/ and that certainly presents a challenge to see if FIM could be used to make similar, and more reproducible, observations.

5. CONCLUSION

In conclusion our calculations indicate that FIM has remarkable potential for determining surface structure if the appropriate experiments, using state of the art techniques, can be performed.

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