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TOWARDS A UNIFIED THEORY OF HELIUM FIELD IONIZATION PHENOMENA

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Abstract - The prediction of the half-width of the helium ion energy distribution is re-examined, in the light of recent progress in imaging theory. It is argued that prediction on the basis of JWKB-type formalisms should be possible: the need is for more realistic models of the electron barrier at a real emitter surface.

1. Introduction

Recent developments in the theory of local-contrast formation in field-ion images have at long last provided it with a more-or-less satisfactory numerical basis. These developments, along with the past history of the local-contrast problem, have recently been reviewed elsewhere [1]. The stimulus for the recent burst of progress was some work of Homeier and Kingham [2]. But from the present author's point of view, the key to improved understanding has been the realisation that the across-surface variations in the average field $F_{av}$ between the critical surface and the emitter surface could be very much greater (as much as 100 times) than the corresponding variations in critical-surface field $F_C$. This turns out to be true even for a relatively close-packed emitter plane, such as tungsten (111). In consequence, it now seems certain that at imaging temperatures near 80K - the local ionization density variations in the critical surface (and hence the observed local contrast) are primarily due to across-surface variations in the ionization (i.e. the electron transition) rate-constant $P_e$.

In contrast theory it is more important to know the relative values of rate-constant at different surface locations than to know the absolute values well. Appropriate relative values can be obtained by the following series of modelling steps:

1. Establishment of a charged-surface model, and calculation of the potential structure above it, - in particular the "one-dimensional potential sections" along lines normal to the surface, passing through critical-surface positions of interest.
2. Fitting, for each such section, between the model surface and the critical surface, an equivalent trapezoidal barrier. This gives the parameters $f$, $B$ and $h$ (or, equivalently, $f$, $B$ and $t$) of the model barrier illustrated in Fig.1(a).
3. Use of some rate-constant formula dependent on these parameters.
4. Normalisation of the resulting rate-constant value, using a divisor given by applying the same rate-constant formula to a reference barrier.

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Figure 1. Models for the electron barrier. (a) Simple trapezoidal barrier, with model parameters shown (solid line). (a) and (b) Reduced potential barrier when coulombic and electron-surface interaction terms are added to the simple barrier (broken line). (b) Triangular barrier used in the GVM approach to modelling the reduced barrier. These diagrams and the others in this paper are schematic.

In connection with Fig.1, we call f the "model-field", h the "step-height" and B the "outer height"; the trapezoidal barrier can be fitted to surface model potentials in more than one way, so a clear distinction must be made between step-height h and surface work-function $\phi$, and between model-field f and the surface fields $F_{av}$ and $F_{cr}$. In the case of inert-gas field ionization the parameter B can be equated to the ionization energy I, though strictly some small interaction terms ought also to be included. For helium ionization on tungsten at best image voltage the reference barrier uses the parameters $B = I = 24.59$ eV; $h = 4.5$ eV; f = 45 V/nm.)

For the first three steps listed above, various alternative choices are available; other modelling procedures are also possible, for example that of Bonn and Good [3]. Some choices of charged-surface model, barrier fitting procedure, and rate-constant formula, are discussed quantitatively in Ref.[1]. It is found that the formula most commonly used in field-ion literature (eq.(3) below) is not fully reliable, but several choice combinations using other formulae give fairly similar results as regards rate-constant variation.

Some conclusions of Ref.[1] relevant to the present discussion are:

1. Quasiclassical theory should be sufficient to describe imaging.
2. In the theoretical derivation of electron transition rate-constant, use of a JWKB or equivalent method is to be preferred, rather than a method based on overlap integrals. This is because it is important to get the wave-functions in the barrier region as near correct as possible [4,5], and this seems to be easier with a JWKB-type formalism than with an overlap-integral formalism. (Past attempts [6,7] to use overlap-integral formalisms to explain local image contrast have been singularly unsuccessful.)
3. In the discussion of local contrast, where relative rate-constant values are the important factor, three-dimensional formalisms do not give significantly better results than one-dimensional formalisms, and the formula - eq.(1) below - derived from a simple trapezoidal barrier will often yield results of adequate quality.

This conclusion, about the pragmatic sufficiency of a one-dimensional JWKB approximation, is at variance with some past thinking. In the mid-sixties, it was the apparent inability of one-dimensional JWKB formalisms to explain the observed spreads for helium field ionization that led to the introduction of overlap-integral formalisms, such as those of Refs.[8,9]; these methods of rate-constant calculation were able to explain observed half-widths.
However, helium field ionization is the same process, whether one is looking at rate-constant variations across the surface or normal to the surface. A necessary objective is thus to look again at all the relevant helium field ionization phenomena, in the hope of eventually developing a coherent theoretical story. In this context, the experimental phenomena that deserve our attention are local contrast, energy-distribution half-widths, and current/voltage characteristics. The first of these is dealt with in Ref.[1] and I concentrate here on energy distribution prediction. This paper should be understood as an interim discussion about what can and cannot be achieved with relatively simple-minded arguments.

2. Rate-constant formulae

As a preliminary, I want to briefly discuss the main rate-constant formulae so far derived, in the context of field-ion imaging, from JWKB-type barrier analyses.

The STB form. The simplest formula is obtained by evaluating a JWKB integral across the simple trapezoidal barrier shown in Fig.1. This gives:

$$P_e = \nu e \exp[-(b/f)(B^{3/2} - h^{3/2})] \quad \text{(STB form)}$$  \hspace{1cm} (1)

where $\nu$ is the "frequency of attempts by the electron to penetrate the barrier", and $b$ is a universal constant related to another universal constant $c_1$ by:

$$c_1 = m_e^{1/2} / \nu = 3.6226 \text{ eV}^{-3/2} \text{ V nm}^{-1} \quad (2a)$$

$$b = (4/3) 21/2 \quad c_1 = 6.8309 \text{ eV}^{-3/2} \text{ V nm}^{-1} \quad (2b)$$

where $m_e$ is the electron mass, $e$ the elementary charge, and $\hbar$ Planck's constant.

The GBM form. Eq.(1) ignores both the atomic potential at the "vacuum" end of the barrier and surface effects at the "emitter" end. Analysis of the triangular barrier of Fig.1(b) attempts to take these into account, and leads to result (3) below, which is attributed to Gomer [10] by Brandon [11] (although it doesn't in fact appear in Ref.[10]), and is rederived by Müller and Tsong [12]. This "Gomer-Brandon-Müller" form, widely used in field-ion literature, is:

$$P_e = \nu e \exp[(-b/f)(B - \hbar)(B - \Delta)] \quad \text{(GBM form)}$$  \hspace{1cm} (3a)

where:

$$\Delta = 2 (c_2 f)^{1/2} \quad (3b)$$

and $c_2$ is another universal constant, defined by:

$$c_2 = (e^3/4\pi e_0) = 1.43998 \text{ eV}^2 \text{ V}^{-1} \text{ nm} \quad (4)$$

The FHG form. A form previously used [13] by the present author is:

$$P_e = a \exp[(-b/f)(\alpha B^{3/2} - \alpha_2 h^{3/2})] \quad \text{(FHG form)}$$  \hspace{1cm} (5)

where $a$ and $\alpha_2$ were presumed to be weakly field-dependent parameters (with $\alpha_2$ of order unity), and $\alpha_2$ is the correction factor proposed by Burgess et al. [14] to the Fowler-Nordheim theory of field electron emission. Values of $\alpha_2$ are tabulated in Ref.[14] as a function of the parameter $y$ given in the present context by:

$$y = 2 (c_2 f)^{1/2} / B \quad \text{(6)}$$

Eq.(12) was developed from the equivalent free-space formula given by Gomer [10]; a broadly similar formula was used by Halpern and Gomer [15] in the context of field ionization into liquid gases.
The HK form. Defects of the above approaches are their one-dimensionality and, more importantly, their lack of attention to the pre-exponential factor. It is relatively straightforward to derive three-dimensional versions of the above formulae (see Ref.[1]), but derivation of an expression for the pre-exponential requires attention to the problems of wave-matching at the atomic end of the barrier. A formula was derived by Goldenfeld [4], but is somewhat unwieldy. More recently Haydock and Kingham [16], using an approximate wave-matching treatment, have produced a result that has the form:

$$P_e = [a_3 f^{1-\zeta} \exp(\zeta)/(B^3/2 - h^3/2)] \exp[-(b/f)(B^3/2 - h^3/2)] \quad (\text{HK form}) \quad (7)$$

where $\zeta$ is a constant, given in terms of the universal constants $c_1$ and $c_2$ defined earlier and an adjustable parameter $Z$ ("the effective charge on the ion") by:

$$\zeta = 2 c_1 c_2 Z / (2B)^{1/2} \quad (8)$$

$a_3$ is a parameter that depends on $B$ and $Z$, but not on $f$ or $h$, and incorporates a numerical adjustment factor; this factor has been so chosen that eq.(7) leads to the known correct (low-field) result for the free-space field ionization of a hydrogen atom. Haydock and Kingham do give a specific form for $a_3$, but it is not clear to the present author that their form is in fact correctly derived, even in the case of inert-gas field ionization, my reservations being about the way the parameter $\nu$ is introduced into their argument. (This difficulty is also present in the derivation of the revised formula, appropriate for metal-ion post-ionisation, given by Kingham in Ref.[17].). However, since $a_3$ does not significantly affect the sensitivity of the rate-constant to field and step-height variations, we need not consider these problems of fine detail here.

Comparisons. A convenient way to compare the performance of the various formulae is to compare values of the partial derivatives $\sigma_f$ and $\rho_h$ defined by:

$$\sigma_f = f \frac{\partial \ln(P_e)}{\partial f} \quad (9)$$

$$\rho_h = \frac{\partial \ln(P_e)}{\partial h} \quad (10)$$

Some results derived for the rate-constant formulae discussed here are shown in Table 1 below. Results are also shown for the three-dimensional version of the FHGa formula, derived in Ref.[1].

<table>
<thead>
<tr>
<th>Rate-constant formula name type</th>
<th>Value of $\sigma_f$ for:</th>
<th>Value of $\rho_h$ for:</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>hydrogen free-space ionization</td>
<td>helium free-space ionization</td>
</tr>
<tr>
<td></td>
<td>$h = 4.5 \text{ eV}$</td>
<td>$h = 4.5 \text{ eV}$</td>
</tr>
<tr>
<td>STB 1D</td>
<td>18.5</td>
<td>18.5</td>
</tr>
<tr>
<td>GBM 1D</td>
<td>16.2</td>
<td>16.0</td>
</tr>
<tr>
<td>FHGa1D</td>
<td>16.7</td>
<td>17.1</td>
</tr>
<tr>
<td>FHGa3D</td>
<td>17.8</td>
<td>18.2</td>
</tr>
<tr>
<td>HKb 3D</td>
<td>17.5</td>
<td>18.0</td>
</tr>
<tr>
<td>true 3D</td>
<td>17.5</td>
<td>n/a</td>
</tr>
</tbody>
</table>

*aTaking $a_2 = 1$ and $d\sigma_2/df = 0$. 
C1D = one-dimensional; 3D = three-dimensional 
Taking Z=1.
These results show that, as far as sensitivity to field and step-height variations are concerned, all the formulae behave in much the same way. In particular, although the 3D formalisms do give better agreement with the true result in the only case which the formulae can be tested, there is no major difference between the results of 1D and 3D formalisms. The GBM formula perhaps looks slightly less reliable than the others.

3. The half-width of the energy distribution

Helium ion energy distribution widths were measured many years ago by Tsong and Muller [18], and more recently by Hanson and Inghram [19]. From these data I take a typical helium half-width (∆E) value to be 0.7 eV, which in the context of an assumed critical-surface field of 45 V/nm implies a space half-width (∆x) of 16 pm. The task is to explain these figures; several approximations and corrections are now investigated.

The basic "uniform-field" model. The simplest approach is to assume a flat surface emitter model, with a uniform field (F_max) above it. In this case, a displacement of the atomic nucleus outwards by a distance 6x causes the step-height of the electron barrier to be reduced by an amount 6h = F_max 6x, as illustrated in Fig. 2; the energy of the departing ion is less by an amount 6E also equal to F_max 6x. For critical-surface ionization the step height is equal to the emitter work-function, assumed 4.5 eV for tungsten. So, from eq.(10), since 6ln(P_e) = 6h, the rate constant falls to half its original value in an energy interval:

$$\Delta E = \ln(2) / \rho_h$$  (11)

Using the STB value for 0h, we obtain ΔE = 1.43 eV. The HK value is slightly less, at 1.35 eV. A similar figure (1.33 eV) was derived in Tsong and Muller's original calculations [18]. It was the discrepancy between this figure and the observed half-widths that, in the mid-sixties, led to the introduction of overlap integral formalisms.

The effect of surface interactions. A defect with the STB, PHG and HK formalisms is that they do not take into account the electron/surface interaction terms in the electron potential energy. The most important of these is the image interaction, and Ref.[1] argues that it can be simulated by using a trapezoidal barrier of slightly greater step-height. In effect, one moves the step out a bit. Ref.[1] estimates that, for critical-surface ionization in the $\phi E = 4.5$ eV case, an increase in step-height of between 3 and 4 eV might be appropriate.

For argument, let us take the effective step-height as 8 eV. The corresponding values of $\rho_h$ are shown in Table 1. (The GBM value is not shown because the GBM formula already incorporates a simulation of electron-surface effects.)
and HK formulae now lead to $\Delta E$ values of 1.08 eV and 1.01 eV respectively; these values are closer to the observed ones, but are still not small enough.

Obviously, this approach is fairly crude, but two points deserve note:

1. With the STB, FHG and HK formulae, the effect of this simulation is to increase the $\rho_h$ value. This is in marked contrast to the GBM formula, where the attempt to simulate electron/surface interactions has led to a reduction in $\rho_h$ as compared with the basic ($h=4.5$eV) STB case. This contrast increases my reservations about the usefulness of the GBM formula.

2. Haydock and Kingham argue [16] that the image interaction can be simulated in their formalism by increasing the size of their "effective charge" $Z$. This will undoubtedly increase the value of $\rho_h$ in a qualitatively correct fashion. But the $\rho_h$-formula derived from their formalism is not $Z$-dependent, so their proposal may not be fully satisfactory.

Field variations near the critical surface. An alternative explanation of the small observed half-widths has been put forward in Ref.[16]. Haydock and Kingham in effect argue that the uniform-field model does not adequately describe the field variation above a protruding surface atom. If, outside the critical surface, the local field $F$ falls off rapidly with distance, then smaller $\Delta x$ (and hence $\Delta E$) values might be expected. They assume field fall-off in the form:

$$F = F^{cr} \frac{R^2}{(R+8x)^2}$$  \hspace{1cm} (12)

where the $R$ is treated as an adjustment parameter. In terms of the present notation, they identify the model-field $f$ with this field $F$ and take $h$ as given by:

$$h = I - eF(x^{cr}+8x)$$  \hspace{1cm} (13)

where $x^{cr}$ is the critical distance between the model surface and the critical surface. They can achieve a $\Delta x$ value of 16 pm if $R$ is taken as about 1.5 nm.

Now this argument has some merit for ionization above a protruding kink-site atom, but for ionization about a flat (111) surface I can see two difficulties with it. First, Haydock and Kingham are in effect fitting trapezoidal barriers by identifying the model-field $f$ with the local field $F$ at the position of the imaging-gas atom nucleus, as illustrated in Fig.3(A). This type of fitting procedure is likely to overestimate the rate at which the barrier increases with distance, and hence may overestimate the rate at which the rate-constant falls off. (This would also be true for ionization above a kink-site atom.)

Second, for ionization above the (111) plane, an $R$-value as small as 1.5 nm does not seem entirely plausible. Calculations carried out using the Forbes and Wafi [20] charge-array model suggest that it is difficult to deduce a plausible $R$-value less than 10 nm, and that the best value might be somewhat higher.

It is thus seems that the Haydock and Kingham calculations do not provide a satisfactory explanation of observed half-widths, certainly not for ionization above facets, and possibly not for ionization above kink-sites.

Average-field variations. An alternative fitting procedure, in which the model field is taken equal to the average field between the step position and the gas-atom-nucleus position, is proposed in Ref.[1]. This procedure is illustrated in Fig.3(B). The model-field again falls off as the gas-atom-nucleus position moves outwards, but the fall-off rate is less than with the fitting method described in the preceding paragraphs. Preliminary calculations, based on the Forbes-Wafi surface model and the STB formula (but ignoring electron/surface interactions) again produce energy half-width estimates of around 1.0 eV. In other words, this approach, too, does not by itself provide a satisfactory explanation of observed half-widths.
Figure 3. Methods of fitting trapezoidal barriers to a non-linear model potential (represented by the broken line). It is assumed that the position of the step has been chosen in some well-defined way. As before, barriers 1 and 2 are the basic trapezoidal barriers applicable for imaging-gas atom nuclei at positions (a) and (b), with position (a) being in the critical surface.

A) Fitting by taking the model-field to be equal to the local field at the gas atom nucleus.

B) Fitting by taking the model-field to be equal to the average field between the position of the gas atom nucleus and the position of the step.

4. Discussion

The following conclusions can be drawn from the above analysis:

1) The "dimensionality" of the JWKB formalism is not a particularly important factor in the context of energy-half-width prediction. In fact, all the JWKB formalisms examined here (with the partial exception of the GBM formula) give similar results.

2) It seems more important (in the context of half-width prediction) to have a good model of the electron barrier than to have a quantum-mechanically "good" formalism.

3) As compared with the simple "uniform field" model, predicted half-width values can be reduced both by using a better model of the field variation above a real surface, and by incorporating electron/surface interactions into the tunnelling barrier.

4) Half-width values as low as those observed have not been satisfactorily predicted by the rather simple analytical methods described here.

Next steps should probably be to use more comprehensive barrier models, and to employ numerical methods to evaluate the corresponding JWKB integrals. At present, I see no reason to doubt that satisfactory explanations of half-widths can be found in the context of one-dimensional JWKB formalisms.

There is also some suggestion in the above analysis that energy half-widths might be different for ionization above a kink-site and above an imaged crystal facet. Whilst there is little indication of this in results so far published, it might be of interest for experiments to look explicitly for such an effect.

REFERENCES