A GENERAL THEORY OF THE CRITICAL VOLTAGE EFFECT

by B. F. Buxton and M. V. Berry

A real-space theory of high energy electron diffraction has been developed by Berry (1971). For systematic diffraction this leads to a one-dimensional eigenvalue problem with a 'projected' potential $\bar{U}(x)$ of period a (Fig. 1). By imposing a Bloch

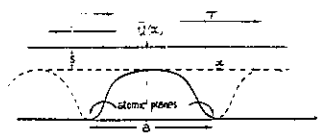

Fig. 1. The 'projected' periodic $\bar{U}(x)$ showing reflection and transmission of waves from an interatomic barrier.

condition with wave number K_0 equal to the x component of the incident electron wavevector k_0, the branches of the dispersion surface $s_j(K_0)$ are the solutions of

$$|T(s)| \cos(K_0 a) = \cos \Phi(s) \quad , \tag{1}$$

where $|T(s)|\exp\{i\,\Phi(s)\}$ is the complex transmission factor from one of the series of 'potential barriers' comprising $\bar{U}(x)$. The zero of energy is chosen at the maxima of $\bar{U}(x)$, and the eigenenergies s_j, labelled in ascending order, are simply related to the wavevectors $k^{(j)}$ of the Bloch waves excited in the crystal.

A degeneracy of the kinetic energies of the Bloch waves thus occurs whenever two values s_j, s_{j+1} coincide. The form of $|T(s)|$ and the $\cos \Phi(s)$ has been discussed by Berry *et al.* (1973), and is shown in Fig. 2. Now $\bar{U}(x)$ scales with the relativistic increase in mass m/m_0 of the incident electrons; therefore a degeneracy of s_n, s_{n+1} may occur if the beam voltage E is chosen so that

$$\left.\begin{aligned} \Phi(a) &= n\pi \\ |T(s)| &= 1 \end{aligned}\right\} \quad , \tag{2}$$

and if we orient the crystal at an appropriate Bragg position

$$K_0 = K_0^B = \pi/a\ (n + 2M) \quad , \tag{3}$$

where M is an integer. The transmission coefficient is unity at a series of resonance energies s which are positive, and a degeneracy occurs if $\bar{U}(x)$ is such that the phase is $n\pi$ at this s, as shown in Fig. 2. Although WKB expressions are good approximations for the one-dimensional eigenfunctions in the regions $s > 0$, and may be used to provide a very good over-all description of the dispersion surface (Steeds and Enfield 1971 and § 5 Berry *et al.* 1973), they cannot include the subtle effects

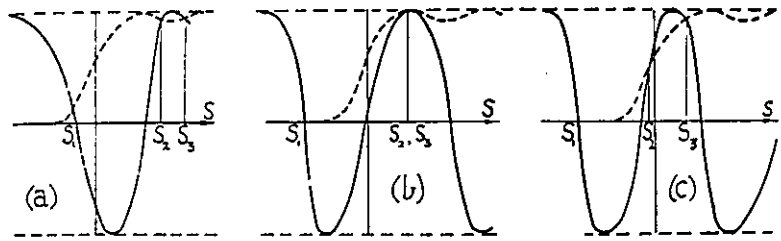

Fig. 2. Curves of cos $\Phi(s)$ (——) and $|T(s)|$ (---) for (a) $E < E_c^{(2)}$; (b) $E = E_c^{(2)}$; (c) $E > E_c^{(2)}$.

which give rise to the resonances. However, we can use the following semiclassical formula for the reflection coefficient, due originally to Landauer,

$$R(s) = \frac{1}{4} \int_{-a/2}^{a/2} dx \quad \frac{d\bar{U}(x)}{dx} \quad \exp\{2i \int_0^x dx' (s-\bar{U}(x'))^{1/2} \quad . \tag{4}$$

This is valid only for $s > 0$, and exhibits a series of zeros (of $R(s)$); since $T^2 + R^2 = 1$, these zeros are the transmission resonances.

Now we introduce a relativistic scaling enabling calculations to be carried out using potentials for $E = 0$ only; define

$$\bar{U}_0(x) = \frac{m_0}{m(E)} \quad \bar{U}(x)$$

$$s_0 = \frac{m_0}{m(E)} \quad s \tag{5}$$

$$\Phi_0(s_0) \equiv \int_{-a/2}^{a/2} dx \sqrt{\{s_0 - \bar{U}_0(x)\}} = \left(\frac{m_0}{m(E)}\right)^{1/2} \Phi(s)$$

then eqns (2) determine the critical voltages from (assuming $\bar{U}(x) = \bar{U}(-x)$)

$$\int_0^{a/2} dx \quad \frac{d\bar{U}_0(x)}{dx} \quad \sin \frac{2n\pi}{\Phi_0(s_0)} \int_0^x dx' \sqrt{\{s_0 - \bar{U}_0(x')\}} = 0 \tag{6}$$

and

$$E_c^{(n)} = m_0c^2 \{(\frac{n\pi}{\bar{\Phi}_0(s_0)})^2 - 1\} \quad . \tag{7}$$

This semiclassical formalism based on Landauer's expression (4) is strictly valid in the asymptotic limit when $\Phi(s)$ and hence n are infinite. However, it is very easy to solve eqns (6) and (7) on a small computer, and we obtain very accurate results for the cases of practical interest of $n = 2 - 6$, for which most critical voltages lie below a few megavolts. For example, the semiclassical second-order critical voltage $E_c^{(2)}$ for Al(200) is 963 kV, while conventional matrix methods give 995 kV. This is one of 50 critical voltages which have been calculated; the average error (which decreases with n) is about 1.5 per cent. Consideration of the integral in (6) indicates that degeneracy of Bloch waves 1 and 2 is impossible for a simple potential such as Al(200) (Fig. 1), and that degeneracy of Bloch waves 3 and 4 is extremely unlikely. Changes in temperature alter the shape of $\bar{U}(x)$ near the atomic planes, and so the critical voltages are very sensitive to the temperature also. Indeed, we have found several pairs of fifth-order critical voltages which approach, coalesce and disappear as the temperature is reduced.

We can also show that, in order to observe a degeneracy of waves n, $(n + 1)$ from a bend contour, the best orientation K_0 is given by (3), with

$$M \approx \frac{1}{2} \frac{a}{\pi} \left(\frac{s_0 m(E_c^{(n)})}{m_0} \right)^{1/2} - n \quad , \tag{8}$$

and the Bragg-reflected dark field is used, corresponding to the reflection $-2K_0^B$.

Rather surprisingly, it is possible, within a very good approximation, to utilize the sensitivity of $E_c^{(n)}$ to the form of $\bar{U}(x)$ to derive an inversion technique to obtain $\bar{U}(x)$ from $E_c^{(n)}$. Introducing the obvious new variable

$$\psi(x) \equiv \frac{2\pi}{\bar{\Phi}_0(s_0)} \int_0^x dx' \sqrt{\{s_0 - \bar{U}_0(x')\}} \quad , \tag{9}$$

(6) can be written as

$$\int_0^\pi d\psi \, f(\psi) \sin n\psi = 0 \quad , \tag{10}$$

which defines $f(\psi)$. If $f(\psi)$ were known, the potential $\bar{U}(x)$ could be obtained exactly as a function of $\psi(x)$. $f(\psi)$ is an odd function of ψ with period 2π, so we may expand it in a Fourier series.

$$f(\psi) = \sum_{n=1}^{\infty} A_m \sin m\psi \quad , \tag{11}$$

and (10) then tells us that at a critical voltage, A_n vanishes. Assuming (for $E_c^{(2)}$), that we may neglect $A_3, A_4, \ldots$, we may determine A_1 from the minimum value of $\bar{U}(x)$ which is related to the *critical angle* K_c/k (§ 3 of Berry *et al.* 1973). Fig. 3 shows a potential 'reconstructed' in this way. Starting from a given potential, K_c was calculated, the potential Fourier-analysed and $E_c^{(2)}$ and $E_c^{(4)}$ were calculated by matrix methods; this gave data for two reconstructions. In terms of the Fourier coefficients of the potential the reconstructions are accurate to about 2 per cent.

In more complicated cases the diffracting planes may not be evenly spaced or there may be more than one atom in a cell unit, so $\bar{U}(x)$ is more complicated. However, knowing the structure, a reconstruction should still be possible and has been carried out for Si(111). Here, Bloch waves 3 and 4 are degenerate at 1145 kV (the semiclassical prediction is 1109 kV), and a fifth-order critical voltage occurs at 1595 kV, which is also the semiclassical prediction. To reconstruct this potential, A_1 and A_2 are fitted to the maximum depth of the potential which is assumed to be at the planes of atoms. A_3 is zero at $E_c^{(3)}$, and A_4, etc. are neglected. The result is still very good as shown in Fig. 4.

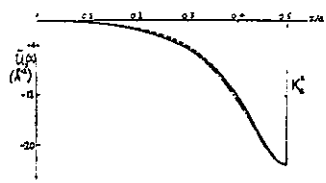

Fig. 3. Comparison of exact (———) and reconstructed potential for Au(200) at 293 K using $E_c^{(2)}$ (----) and $E_c^{(4)}$ (.-.-.-.)

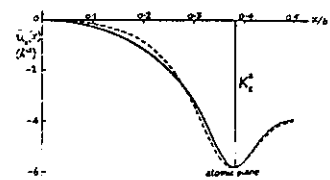

Fig. 4. Comparison of exact (———) and reconstructed (---) potentials for Si(111) at 293 K.

The semiclassical methods outlined here will be elaborated in a more general and detailed discussion of the degeneracy of Bloch waves in systematic high energy electron diffraction to be published later.

References

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