

Image potential effects for low and high energy electrons

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Developments in the theory of the image potential of charged particles moving near condensed matter surfaces are sketched. Some applications to electrons, (i) localized in image potential states, (ii) tunneling across a gap between two metals, (iii) localized in fast microprobe beams, are considered.

1. Introduction

The complex potential resulting from the polarization induced when a charged particle approaches a solid corresponds to the self-energy of the incident particle due to its interaction with a large variety of modes. Electron-hole pairs, bulk plasmons, phonons and surface plasmons may contribute to the response of the medium. Hence the self-energy is the result of the virtual excitation of all possible modes.

When the particle is far away from the surface it couples to long-wavelength collective modes of the system. The importance of such modes to the image potential was emphasized independently by a number of authors [1]. This followed experimental evidence of deviations from classical image potentials and attempts to find semi-empirical corrections [2] on the one hand and the earlier discovery that van der Waals forces also have their origin in surface modes on the other [3].

Several workers addressed the problem of dynamical correction to the real part of the semiclassical surface potential without including dispersion of the surface modes [4]. The imaginary part of the semiclassical image potential was also studied [5]. The q dependence of the relevant modes becomes important at distances less than a few screening lengths from the surface. The effect of mode dispersion on the real part of the potential has been studied by several workers [6].

Theories including the quantal properties of the external particle have been proposed [7]. Flores and Garcia-Moliner [8] derived a formula for the complex self-energy, valid for an arbitrary dielectric function both within a semiclassical and a quantum formulation. A surface-plasmon-pole approximation [9] could

in principle incorporate many of the recent developed aspects of the interaction [10] and still lead to manageable expressions for the potential, for use in energy loss, low-energy electron diffraction (LEED), reflection high-energy electron diffraction (RHEED), and ion-scattering calculations.

Manson and Ritchie [11] have presented a general expression for the space-dependent, complex self-energy of a projectile interacting with a specified N -particle target. The quantal self-energy was obtained from high-order perturbation theory including full three-dimensional effects.

In the region where the momentum dispersion of the excitations becomes important local effects associated with the tail of the electronic charge density penetrating into the vacuum [12] should be taken into account. This complicates the calculation since one needs to include at the same time local and non-local effects. Attempts by Ossicini and Bertoni [13] to introduce non-locality in the functional density method in order to reproduce the asymptotic form of the image potential have been criticized by several authors [14,15]. The barrier potential for tunneling electrons at a metal-metal interface has been obtained as a sum of a local and a non-local contribution, by de Andres et al. [16].

The growing interest in the scanning transmission electron microscope STM [17] has revived experimental and theoretical work directed toward understanding the influence of the dynamical image potential on tunneling [18–23]. A study of the dependence on the interface potential of the oscillations of tunneling conductance with applied bias voltage observed in STM has been presented recently [24].

At short distances from the surface, dispersion effects within the non-local theory and local effects as well, become important. Non-linear screening and the breakdown of translational invariance in the case of real surfaces also become important. This is particularly important in the case of image states at metal surfaces [25], where a detailed knowledge of the attractive potential is necessary [25–27].

Predictions of energy loss at metal surfaces as a function of the impact parameter [5] have been subject to experimental tests due to recent developments in the scanning transmission electron microscope (STEM) which have made it possible to study electronic excitations of inhomogeneous systems in highly localized regions. This is achieved by recording changes in energy loss distributions measured when a well-focused (~ 0.5 nm) probe of swift (~ 100 keV) electrons is scanned across a specimen [28]. The probability of energy loss is directly related to the imaginary part of the self-energy of the electrons while the real part is related to the deflection force or image force experienced by an electron beam when moving in the neighbourhood of a surface [29].

In this paper we shall first be concerned with STEM near metal surfaces of different geometries. The role of the dynamical image potential in STM and the importance of the different variables such as screening, tunneling times or electron recoil in the tunneling process will be analyzed next. Finally we shall address the question of image states at metal surfaces. In all of the above the dynamical image potential is a central concept.

2. Scanning transmission electron microscopy (STEM)

In the case of scanning transmission electron microscopy (STEM) much information is contained in the impact parameter dependence of the interaction. At the high energies of the STEM electrons and for impact parameters greater than a few screening lengths, recoil effects and the Q dependence of the surface response function may be neglected. The characteristic length involved in these problems is $\lambda_e \sim v/\omega_s$, thus the interaction is weaker the greater the velocity but one has to go further into vacuum in order to reach the classical asymptote. This is illustrated by the following expression for the imaginary part of the self-energy for an electron moving at a distance z from a planar surface whose response is described in terms of an undispersed surface plasmon with eigen frequency ω_s [5];

$$\Sigma_i(z) = -(\omega_s/2v)K_0(2\omega_s z/v), \quad (1)$$

where v is the electron velocity and K_0 is a modified Bessel function of the second kind.

In electron microscopy, it is of interest to express the retarding force acting on the particle in terms of the probability of energy loss $P(\omega)$ as

$$S = \int_0^{\infty} P(\omega) \omega \, d\omega. \quad (2)$$

Then $P(\omega)$ may be written, for a particle moving outside a planar surface of a solid that is characterized by the local response function $\epsilon(\omega)$, as [5,28].

$$P(\omega) = \frac{2}{\pi v^2} \operatorname{Im} \left(\frac{\epsilon - 1}{\epsilon + 1} \right) K_0(2\omega z/v). \quad (3)$$

The properties of the Bessel function K_0 give rise to interesting behaviour in the spectra. For large values of its argument, $x = 2\omega z/v$ it falls off like e^{-x}/\sqrt{x} and the $P(\omega)$ can extend to distances of the order of (v/ω) , which are typically 50 Å in the valence loss region. On the other hand, at small values of x (but still greater than a few screening lengths), the function has a rapid logarithmic variation so the losses at larger ω , in particular, can vary rapidly with impact parameter thus allowing a high spatial resolution that may explain recent experimental findings [30].

Inside the solid ($z < 0$) one has

$$P(\omega) = \frac{2}{\pi v^2} \left\{ \operatorname{Im} \left(-\frac{1}{\epsilon(\omega)} \right) \ln \left(\frac{q_c v}{\omega} \right) + \left[\operatorname{Im} \left(-\frac{2}{1+\epsilon} \right) - \operatorname{Im} \left(-\frac{1}{\epsilon} \right) \right] K_0(2\omega |z|/v) \right\}. \quad (4)$$

The logarithmic term yields the ordinary loss rate to volume excitations, q_c is a cut-off wave number, and the terms containing the K_0 function describe boundary corrections to these losses.

In the case of spherical particles of radius a the probability of losing energy ω at impact parameter $b > a$ is given by [31,32].

$$P(\omega) = \frac{4a}{\pi v^2} \sum_{L=0}^{\infty} \sum_{m=0}^L \frac{2 - \delta_{0m}}{(l+m)!(l-m)!} \left(\frac{\omega a}{v} \right)^{2L} K_m^2(\omega b/v) \operatorname{Im} \left(\frac{L\epsilon(\omega) - 1}{L\epsilon(\omega) + L + 1} \right). \quad (5)$$

Here δ_{0m} is the Kronecker delta, and K_m is the modified Bessel function of the order m . In many experimental situations $b \sim a$ and one needs to include many L 's in eq. (5).

3. Tunneling

Jonson [19] investigated the dynamical image potential appropriate for tunneling electrons through a rectangular barrier by way of the Green function formalism. He found deviations from calculations based on the static image potential whenever $(z/k) \gg \omega_S^{-1}$ where z is the distance from the first surface, E is the energy of the particle and $k = [2(E - V(z))]^{1/2}$. This is for traversal times shorter than the time associated to the screening response of the plasmon field. Echenique et al. [21] performed a calculation of the dynamic image potential for a very simple but complete basis set of eigenfunctions of the one-electron potential experienced by the electron in the non-interacting system. They choose a set of eigenfunctions corresponding to the δ -function one-electron potential located at the surface and assume that the dynamical properties of the metal which is located in the half-space $z < 0$, may be represented by surface plasmon oscillators. The z -dependent self-energy is given by,

$$\Sigma(z) = \sum_{n=0}^{\infty} \sum_Q \frac{\alpha_Q^2 e^{-Qz}}{\epsilon_0 - \epsilon_n + \frac{1}{2} [P^2 - (P - Q)^2] - \omega_{SQ} + i\delta} \left(\frac{U_n(z)}{U_0(z)} \int dz' U_0(z') e^{-Qz'} U_n^*(z') \right). \quad (6)$$

Here α_Q is the coupling constant in the electron-surface plasmon interaction Hamiltonian,

$$\alpha_Q^2 = \frac{\pi\omega_S^2}{AQ\omega_{SQ}}, \quad (7)$$

A is a normalization area,

$$\omega_S = \lim_{Q \rightarrow 0} \omega_{SQ},$$

and ω_{SQ} has the surface-plasmon-pole type of behaviour [9]. P is the momentum perpendicular to the z -axis of the electron in its initial state, and ϵ_n is the eigenenergy associated with perpendicular motion corresponding to the state function $U_n(z)$.

Using the complete, orthonormal basis set of the δ -function potential, the explicit expression for the z -dependent self-energy for $P = 0$, and in the dispersionless limit, is found to be [21]

$$\Sigma_0(z) = -\frac{\omega_S}{2} \int_0^\infty dQ \frac{e^{-2Qz}}{\omega_S - \lambda_0 Q} \left(1 - \frac{Q \exp\left\{-\left[(2\omega_S + Q^2 + \lambda_0^2)^{1/2} - \lambda_0 - Q\right]z\right\}}{(2\omega_S + Q^2 + \lambda_0^2)^{1/2} - \lambda_0} \right). \quad (8)$$

Here $\lambda_0^2/2$ is the energy measured from the top of the barrier. Expression (8) tends to the well-known classical image potential at large distances from the surface, $-1/4z$, for any value of the strength λ_0 of the δ -function potential. At the apparently singular point $\omega_S = \lambda_0 Q$ the expression in square brackets in eq. (8) vanishes, and, as one expects for $P_0 = 0$, there is no imaginary part of $\Sigma(z)$. The self-energy obtained by Jonson when the electron is inside a rectangular barrier is identical with the first term of eq. (8). Inclusion of recoil terms eliminates the unphysical divergence at $\omega_S = \lambda_0 Q$ and an spurious imaginary term in the self-energy obtained by Jonson. The results of Echenique et al. corroborate the main physics in Jonson's analysis. Similar results have been obtained by Sols and Ritchie [31] and Sunjic [32] within the self-energy formalism for a two plane geometry.

Büttiker and Landauer [33] have shown that in the case of tunneling through a square barrier of width L whose height V_0 is modulated at a frequency ω , the component of the tunneling current is governed by the mean square barrier only when a characteristic traversal time τ_0 , defined when polarization is neglected, as

$$\tau_0 = L/v = L/\sqrt{2V_0}$$

is much greater than the inverse of the modulating frequency ω , thus confirming the deviations predicted by Jonson in the case of $\omega_S \tau_0 \leq 1$.

In the case of STM the modulation, studied by Büttiker and Landauer is generated by the tunneling electron and therefore self-consistency in the determination of effective barrier and is a key ingredient of the problem [32]. Persson and Baratoff [22] have discussed the self-consistent determination of the dynamic polarization or "image potential" in tunneling within linear response theory in a non-perturbative manner via the "instanton" approximation [34]. Persson and Baratoff find that in all cases the contribution of the dynamical image potential to the tunneling exponent decreases monotonically for decreasing $\omega_S \tau_0$. They estimate that using the static image potential in calculating the tunneling exponent in a typical STM experiment [35] overestimates the effect of the barrier lowering on the exponent by 20–30%. In addition Persson and Baratoff find that if $\omega \tau_0$ is sufficiently small the tunneling exponent and hence the effective barrier will remain finite even if the barrier calculated using the static image potential collapses to zero.

In all the previous models, the crucial approximations are twofold: (i) the tunneling electron is assumed to interact only with the surface plasmon field; (ii) the electronic density in the interface region between the two surfaces is neglected. In a more complete approach, local effects associated with the interface

electronic charge should be included; in other words, the local exchange and correlation potential, as calculated in a LD approximation, yields important contributions for the distances of interest (10–15 Å) in the STM. Ferrante and Smith [36] have shown that the electron density of a metal–metal interface is practically the superposition of the charges of the two independent surfaces. Using this approximation, de Andres et al. [16] have found a substantial contribution to the interface potential barrier due to the overlap of the metal electronic charges. This agrees with more detailed LDA calculations [37], and shows that in a complete approach to the calculation of the interface potential barrier, local (as given by the local metal charges) and non-local effects (as discussed above using the surface plasmon field) should be both included.

These two independent approaches have been followed to solve this problem. In one case, the LDA has been supplemented with a prescription to calculate non-local potentials [38,39] and in the other, a self-energy approach has been used to calculate the interface potential [16,40].

The first approach has been recently criticized by Tarazona et al. [14] and, although Bertoni et al. [41] have shown the method to give good estimations of the non-local corrections for short metal–metal interface distances, it does not give the appropriate non-local corrections to the interface potential for intermediate and long distances ($d \geq 5$ Å).

The WDA formulation by Gunnarsson and Jones [42] gives the correct image behavior of the exchange and correlation potential, V_{xc} , but as noted by Tarazona and Chacon [14,15] the values of the image plane position are unrealistically negative. This is due to the model of the exchange–correlation hole used in the methods, which is good when the electron out of the surface is not far from it (≤ 5 Å), but extends too much inside the surface when the electron is far away. However if the position of the image plane z_0 is defined as the centre-of-mass of the induced charge responding to an external field, z_0 the results obtained in this way are different than the ones obtained from the asymptotic behavior of V_{xc} with WDA results only slightly less than the corresponding LDA ones. The same trend exists if one starts from a self-energy approach [43] to calculate exchange and correlation. The results of Eguiluz et al. [43] are of the same type as the ones obtained in ref. [41] from V_{xc} .

In the second approach, de Andres et al. [16] have shown how to use the complete self-energy approach to obtain the local and non-local effects to the interface potential. Local effects are extracted for Ferrante et al.'s approach while non-local effects are obtained from the interface image potential once the "local" effects included in this potential due to the electronic local charge, are subtracted out. The main results coming out of this calculation are the following: (i) non-local effects are small for distances smaller than 5 Å; (ii) non-local effects dominate the interface potential barrier for distances larger than 10 Å; (iii) in the intermediate region, $5 \text{ Å} < d < 10 \text{ Å}$, both local and non-local effects contribute.

Pitarke et al. [41] have used these calculations to obtain the apparent barrier height of a Au–W interface, with the apparent barrier defined by

$$\phi_{ap} = \frac{1}{8} \left(\frac{d \ln I}{ds} \right)_{V_a}^2,$$

where I is the STM tunneling current intensity for the metal–metal interface as a function of the separation s for a given applied constant bias, V_a . We believe that these are the most complete results for a metal–metal interface as they include both local and non-local effects. Several comments are worthwhile: (i) first, the apparent barrier height is larger than the mean barrier height and even larger than the maximum barrier height; (ii) the apparent barrier height is always smaller than the average workfunction, although it tends to this limiting value for large separations; (iii) in fact, the apparent barrier is very close to the average workfunction for $d > 6$ Å when local and non-local effects are still important (Lang, Binnig et al.). Notice that in this case the mean barrier height is substantially smaller than the average workfunction. All these results show the importance of including local and non-local effects, in obtaining the mean barrier height and the tunneling intensity. The apparent barrier height is, however, almost

constant for distances larger than 6 Å, explaining why experiments show for the intermediate or large distances region an almost linear $\ln I$ versus s characteristic.

4. Image states

If the energy dependence of the denominator of eq. (6) is neglected the sum over “ n ” can be eliminated by means of the closure approximation and one recovers the expression for the semiclassical image potential. Several calculations of binding energies and effective masses of image states using the self-energy formalism have been performed showing that the binding energy within the infinite barrier surface plasmon model, decreases with decreasing density while the effective mass correction increases with decreasing density, but remaining within 5% for the material studied [44]. Density functional calculations [45] of binding energy and calculations going beyond second order [46] in the self-energy show the same tendency.

In metal surfaces inclusion of scattering by the crystal is crucial to understand the details of the binding. The variation of the WKB phase of the classical image potential reflectivity with energy $\phi_{\text{WKB}} \sim 1/\sqrt{-E}$ (energy measured with respect to the vacuum level) leads to a Rydberg series [47] which for a constant phase representing the crystal reflectivity ϕ_c gives the following expression for the binding energy

$$E_b = -\frac{1}{32(n+a)^2}; \quad a = \frac{1}{2}(1 - \phi_c/\pi).$$

In most experimental cases ϕ_c varies from 0 at the bottom of the gap to π at the top. If the vacuum level occurs at the top of the gap, as it occurs in the (111) faces of some fcc metals, then $\phi_c = \pi$, $a = 0$ and $E_b = -0.85 \text{ eV}/n^2$. In the (100) faces of the same fcc metals quoted above, $\phi_c = \pi/2$, $a = 0.25$ and $E_b = -0.85/(n+0.25)^2$. Therefore one expects that for such fcc metals the image states would be more tightly bound in the (111) than in (100) faces. This has been found experimentally [48]. The most striking aspect of the image potential is that it leads to a Rydberg series of bound states.

As the energy normal to the surface approaches the vacuum level, the states, due to the classical image potential tail, get closer in energy $\Delta E_n \sim 1/n^3$ but at the same time the center of gravity of the resonant state charge distribution moves further away from the surface. This leads to a smaller probability of coupling to surface excitations, hence the energy broadening of the levels becomes smaller, allowing the Rydberg series to be resolved in some cases [47]. Model calculations of lifetimes of the first image states at the surfaces of Cu and Ag [49,50] are in good agreement with recent experimental observations [51,52]. Very recently, Shoenlein et al. [53] have reported the first time-resolved studies of higher order image potential states on Ag(100). Their results are in agreement with the predicted theoretical scaling [54].

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