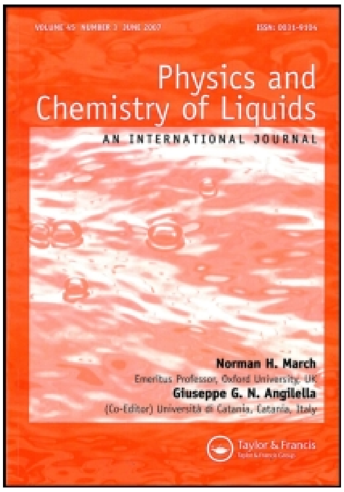

Physics and Chemistry of Liquids
Publication details, including instructions for authors and subscription information:
http://www.informaworld.com/smpp/title~content=t713646857

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To cite this Article Amovilli, C. , March, N. H. and Pfalzner, S.(1991) 'Model of Confined Atoms in Arbitrary Static Electric Fields: Relevance to Non-degenerate Plasmas', Physics and Chemistry of Liquids, 24: 1, 79 — 89
To link to this Article: DOI: 10.1080/00319109108030651
URL: http://dx.doi.org/10.1080/00319109108030651

Phys. Chem. Liq., 1991, Vol. 24, pp. 79–89
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MODEL OF CONFINED ATOMS IN ARBITRARY STATIC ELECTRIC FIELDS: RELEVANCE TO NON-DEGENERATE PLASMAS

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(Received 2 February 1991)

The inhomogeneous electron cloud in atomic ions 'confined' in hot plasmas and subjected to high static electric fields is studied, because of a body of experimental data on multiphoton ionization. In particular, the canonical (Bloch) density matrix is obtained in closed form for independent electrons moving in a static electric field of arbitrary strength and confined by a harmonic oscillator potential. To bring the model into contact with atoms in plasmas, the oscillator force constant is connected with the plasma density. For non-degenerate electrons an 'atomic' potential is included, by means of the Thomas–Fermi (TF) method. In an Appendix, a fully non-local theory is then developed which transcends this TF approximation. Simple numerical examples are presented for realistic values of field, temperature and plasma density.

KEY WORDS: Dilute plasmas, Strong electric field, Inhomogeneous electron cloud.

1 INTRODUCTION

Recently, the Thomas–Fermi method has been used, together with some refinements, to discuss atoms in cold dense plasmas[1]. The present paper is in a related area, though the emphasis now is on atomic ions 'confined' in hot plasmas, and subjected to high static electric fields. Various models already exist[2]; the long-term aim must be to bring such models into contact with a body of experimental data on multiphoton ionization. Characteristic of such experiments are (i) low ionic densities and (ii) strong electric fields.

The present work has been motivated, in part, by the very recent study of Brewczyk and Gajda[3], who applied the Thomas–Fermi (TF) method to such systems. One of us[4] has subsequently used the same approach to study numerically the dependence of the self-consistent atomic potential on electric field.

The aim of the present investigation may be summarized as follows:

i) To present a soluble model for a confined assembly of independent electrons subjected to a static electric field of arbitrary strength E. The confinement is achieved by imposing a harmonic restoring force, in addition to the electric field.

ii) To relate to atomic ions in hot, non-degenerate plasma.

The outline of the paper is a presentation first in section 2 of the solution of the Bloch equation for the canonical density matrix $C(\mathbf{r}, \mathbf{r}_0, \beta, E, \omega)$ for independent electrons in a constant electric field E, with harmonic restoring force corresponding to an oscillator angular frequency ω. Here β is the reciprocal of the thermal energy k_BT. In the first part of section 2, the electric field is taken as the z axis. Then this solution is readily generalized to include harmonic restoring forces also in the x and y directions.

Section 3 is then concerned with relating the above model to atomic ions in a hot, non-degenerate plasma in an external electric field. The first step is to add an 'atom-like' potential $V(\mathbf{r})$ to the model solved in Section 2. Strictly $V(\mathbf{r})$ should be calculated self-consistently as a function of β, E and the plasma density. While this is not attempted here, a model potential $V(\mathbf{r})$ is incorporated into the treatment of Section 2 by the TF approximation. The second step is to connect the strength of the harmonic potential with the plasma density.

Section 4 presents some typical numerical examples, for realistic values of field, temperature and plasma density. A summary, with some suggestions for future work, concludes the body of the paper. However, in a substantial Appendix, a non-local theory is developed, for non-degenerate electrons moving in a model potential $V(\mathbf{r})$, which transcends the TF approximation.

2 SOLUTION OF BLOCH EQUATION FOR CANONICAL DENSITY MATRIX FOR HARMONIC FORCE CONFINEMENT AND IN STATIC ELECTRIC FIELD

The starting point of this study is to assume that independent electrons move in a static electric field of arbitrary strength E, added to which is a confining harmonic oscillator potential, corresponding to angular frequency ω. The method employed is then to construct the canonical density matrix $C(\mathbf{r}, \mathbf{r}_0, \beta, E, \omega)$, by solution of the Bloch equation

$$\hat{H}_{\mathbf{r}} C = -\frac{\partial C}{\partial \beta}. \tag{2.1}$$

From the definition of C in terms of the one-electron wavefunctions $\psi_i(\mathbf{r})$ of the

Hamiltonian $\hat{H}$, and the corresponding eigenvalues ϵ_i, namely

$$C(\mathbf{r}, \mathbf{r}_0, \beta) = \sum_i \psi_i(\mathbf{r})\psi_i^*(\mathbf{r}_0) \exp(-\beta\varepsilon_i), \tag{2.2}$$

it follows from the completeness theorem for eigenfunctions that C must satisfy the 'boundary' condition

$$C(\mathbf{r}, \mathbf{r}_0, \beta = 0) = \delta(\mathbf{r} - \mathbf{r}_0). \tag{2.3}$$

The rest of this section is devoted to solving Eq. (2.1), with condition (2.3) for $\hat{H} = \hat{H}_0 - eEz +$ *harmonic confining potential energy*. Here $\hat{H}_0$ is the free particle Hamiltonian $-\hbar^2/2m\nabla^2$. In atomic units ($e = 1, m = 1, \hbar = 1$), the canonical density matrix for free particles, satisfying Eqs. (2.1) and (2.3) with $\hat{H}$ replaced by $\hat{H}_0$ is

$$C_{free} = \frac{1}{(2\pi\beta)^{3/2}} \exp\left(-\frac{|\mathbf{r} - \mathbf{r}_0|^2}{2\beta}\right). \tag{2.4}$$

Evidently, the solution of the model problem posed above must reduce to Eq. (2.4) when $E = 0$ and when the harmonic restoring force is switched off. Let us immediately exploit the form (2.4) which is a product of x, y and z terms having the structure

$$\frac{1}{(2\pi\beta)^{1/2}} \exp\left(-\frac{(x - x_0)^2}{2\beta}\right),$$

for the case when the harmonic restoring force, represented by potential energy $\frac{1}{2}m\omega^2z^2$, confines electrons only along the field direction, namely the z axis.

2.1 *Confining harmonic force only along field direction*

Motivated by the form (2.4), one can write, since the motion in x and y directions is unaffected:

$$C(\mathbf{r}, \mathbf{r}_0, \beta, E, \omega) = \frac{1}{2\pi\beta} \exp\left(-\frac{(x - x_0)^2}{2\beta} - \frac{(y - y_0)^2}{2\beta}\right) C_z(z, z_0, \beta, E, \omega). \tag{2.5}$$

Evidently the differential equation for C_z on the right-hand side (2.5) can be readily obtained. The point to be stressed is that the potential terms in the Hamiltonian $\hat{H}$ can be rearranged as

$$\tfrac{1}{2}m\omega^2z^2 - eEz = \tfrac{1}{2}m\omega^2\left[z - \frac{eE}{m\omega^2}\right]^2 - \frac{e^2E^2}{2m\omega^2} \tag{2.6}$$

Thus, one has to deal with a harmonic oscillator with a shift of origin proportional to electric field. Using the study of Stephen and Zalewski[5], following the earlier work of Sondheimer and Wilson[6] on the free electron diamagnetism, one can show after some calculation that the form of C on the right-hand side of Eq. (2.5) is

$$C_z(z, z_0, \beta, E, \omega) = \left[\frac{\omega}{2\pi \sinh(\beta\omega)}\right]^{1/2} \times \exp\left(-\frac{\omega}{4}\tanh\left(\frac{\beta\omega}{2}\right)\left(z + z_0 - \frac{2E}{\omega^2}\right)^2 - \frac{\omega}{4}\coth\left(\frac{\beta\omega}{2}\right)(z - z_0)^2\right) \times \exp\left(\frac{\beta E^2}{2\omega^2}\right). \quad (2.7)$$

If the limit $\omega \to 0$ is taken in Eq. (2.7) and the result is inserted into Eq. (2.5) then one obtains

$$C_{\omega\to 0} = \frac{1}{(2\pi\beta)^{3/2}} \exp\left(-\frac{|\mathbf{r} - \mathbf{r}_0|^2}{2\beta} + \frac{\beta E}{2}(z + z_0) + \frac{\beta^3 E^2}{24}\right) \quad (2.8)$$

which was given earlier by Harris and Cina[7]. It can be directly verified by insertion of Eqs. (2.7) and (2.8) into the appropriate forms Bloch Eq. (2.1) that they are solutions, and the limit $\beta \to 0$ in each is readily shown to satisfy Eq. (2.3).

Plots will be given below, in section 4, of C_z in Eq. (2.7) on the diagonal $z_0 = z$ for realistic values of β, E and ω, the last of these being connected with the plasma density in Section 3 below. However, before turning to that, the interest in atomic ions confined in plasma means that the electronic motion should be confined also in the x and y directions. Since there is axial symmetry around the field direction, this necessitates the introduction of only one further force constant or equivalently a further frequency which will be denoted by ω_1.

2.2 *Additional harmonic force confinement in x and y directions*

Using the results of Refs [4] and [5], it is a straightforward matter to introduce the new potential energy contribution $\frac{1}{2}m\omega_1^2(x^2 + y^2)$ into the free-particle terms in x and y on the right-hand side of Eq. (2.5). Then the new form of Eq. (2.5) reads

$$C(\mathbf{r}, \mathbf{r}_0, \beta, E, \omega, \omega_1) = \left[\frac{\omega_1}{2\pi \sinh(\beta\omega_1)}\right] \times \exp\left(-\frac{\omega_1}{4}\tanh\left(\frac{\beta\omega_1}{2}\right)(x + x_0)^2 - \frac{\omega_1}{4}\coth\left(\frac{\beta\omega_1}{2}\right)(x - x_0)^2\right) \times \exp\left(-\frac{\omega_1}{4}\tanh\left(\frac{\beta\omega_1}{2}\right)(y + y_0)^2 - \frac{\omega_1}{4}\coth\left(\frac{\beta\omega_1}{2}\right)(y - y_0)^2\right) \times C_z(z, z_0, \beta, E, \omega) \quad (2.9)$$

Again representative plots of Eq. (2.9) on the diagonal $\mathbf{r} = \mathbf{r}_0$ will be presented in Section 4.

This is the point to turn to the way one now introduces the atomic ion, which is modelled through a suitable one-body potential $V(\mathbf{r})$. In general, self-consistent determination of $V(\mathbf{r})$ in a plasma will lead to the potential depending not only on $\mathbf{r}$ but also on temperature and electric field. Though such self-consistency is not attempted here, some discussion will be given in Section 4 of the regime where the field dependence of $V(\mathbf{r})$ might be unimportant.

3 INTRODUCTION OF MODEL POTENTIAL ENERGY $V(\mathbf{r})$ REPRESENTING ATOMIC ION

Let us now turn to the problem of switching on a model potential $V(\mathbf{r})$ to the Hamiltonian used in Section 2. Denoting the canonical density matrix calculated there by $C^{(0)} \equiv C(V=0)$, the simplest approximation is to follow the ideas of the TF method. Then, with slowly varying $V(\mathbf{r})$ for which the assumptions of this approximation are valid, one can return to the definition (2.2), and simply move all eigenvalues ϵ_i by the same (almost constant—) amount $V(\mathbf{r})$, the wavefunctions $\psi_i(\mathbf{r})$ being unaffected to the same order of approximation. Hence one can write for the diagonal form of the canonical density matrix

$$C(\mathbf{r}, \beta) = C^{(0)} \exp(-\beta V(\mathbf{r})). \tag{3.1}$$

It is relevant to the discussion of Section 2 to note that if V were simply the electric field term $-eEz$ in Eq. (2.6) and this was switched on the free particle form (2.4), then the additional factor multiplying $C^{(0)}$ would be $\exp(\beta Ez)$. This is precisely the factor present in the diagonal form of Eq. (2.8). However, potentials $V(\mathbf{r})$ in atomic ions evidently have Coulomb singularities at nuclei, so that Eq. (3.1) is a less favourable approximation in this case than for the linear potential $-eEz$.

3.1 *Transcending Thomas–Fermi approximation*

As proposed by Hilton *et al.*[8], one can contemplate generalizing the form (3.1) by writing

$$C(\mathbf{r}, \beta) = C^{(0)} \exp[-\beta U(\mathbf{r}, \beta)] \tag{3.2}$$

where the so-called effective potential U now becomes a function of β, even if the model potential $V(\mathbf{r})$ is chosen to be independent on temperature. Hilton *et al.* propose then to calculate U to first-order only in V. To illustrate their results, if $C^{(0)}$ is replaced by the free-particle limit in zero field, then the first order term of U, say U_1, can be written explicitly in the non local form

$$U_1(\mathbf{r}, \beta) = \int d\mathbf{r}_1 V(\mathbf{r}_1) G_0(\mathbf{r}, \mathbf{r}_1, \beta) \tag{3.3}$$

where

$$G_0(\mathbf{r}, \mathbf{r}_1, \beta) = \frac{1}{\pi\beta|\mathbf{r}_1 - \mathbf{r}|} \exp\left[-\frac{2(\mathbf{r}_1 - \mathbf{r})^2}{\beta}\right]. \tag{3.4}$$

In the Appendix, Eq. (3.3) is generalized to apply to switching $V(\mathbf{r})$ on to the model problem of Section 2. If one restricts oneself here to Eq. (3.3), Hilton *et al.* plot $U_1(\mathbf{r}, \beta)$ for various cases: the Coulomb singularity at $\mathbf{r} = 0$ is removed by the non-local form (3.3) for any finite β.

3.2 *Connection of harmonic confining force constants and frequencies with plasma density*

One application of the above described model is to the modelling of plasmas. In the statistical description of dense plasmas it is a common method to estimate the radius of the cell occupied per atom by dividing the volume by the number of particles

$$r = \left(\frac{4\pi n_i}{3}\right)^{-1/3}, \tag{3.4}$$

where n_i is the ion number density. In the case of harmonic confining forces, the radius of this cell can be set equal to the wavelength of the harmonic force. This boundary is, unlike that in the Thomas–Fermi model, a smooth well because of the harmonic potential. The advantage of this boundary is that the electrons are not totally fixed in their cell but some tunneling is allowed as well. So the connection between the frequency of the harmonic force and the plasma density is given by

$$\omega = \pi\left(\frac{4\pi n_i}{3}\right)^{1/3}. \tag{3.5}$$

This definition of the force constant will be employed below in some illustrative examples. The division of the volume of the plasma into these small cells is best applicable in the case of dense plasmas. The model described above is restricted in the density range because of the assumption of a non-degenerate plasma, but using Fermi–Dirac statistics instead of Maxwell–Boltzmann the range of applicability of this approach could be widened to embrace very high densities ($\sim 10^{23}$ particles per cc).

4 SIMPLE ILLUSTRATIVE EXAMPLES

Here, some numerical examples will be presented. As far as possible, bearing in mind the limitations of the model, the examples are designed for conditions which can be achieved in laboratory experiments. However only non-degenerate plasmas will be

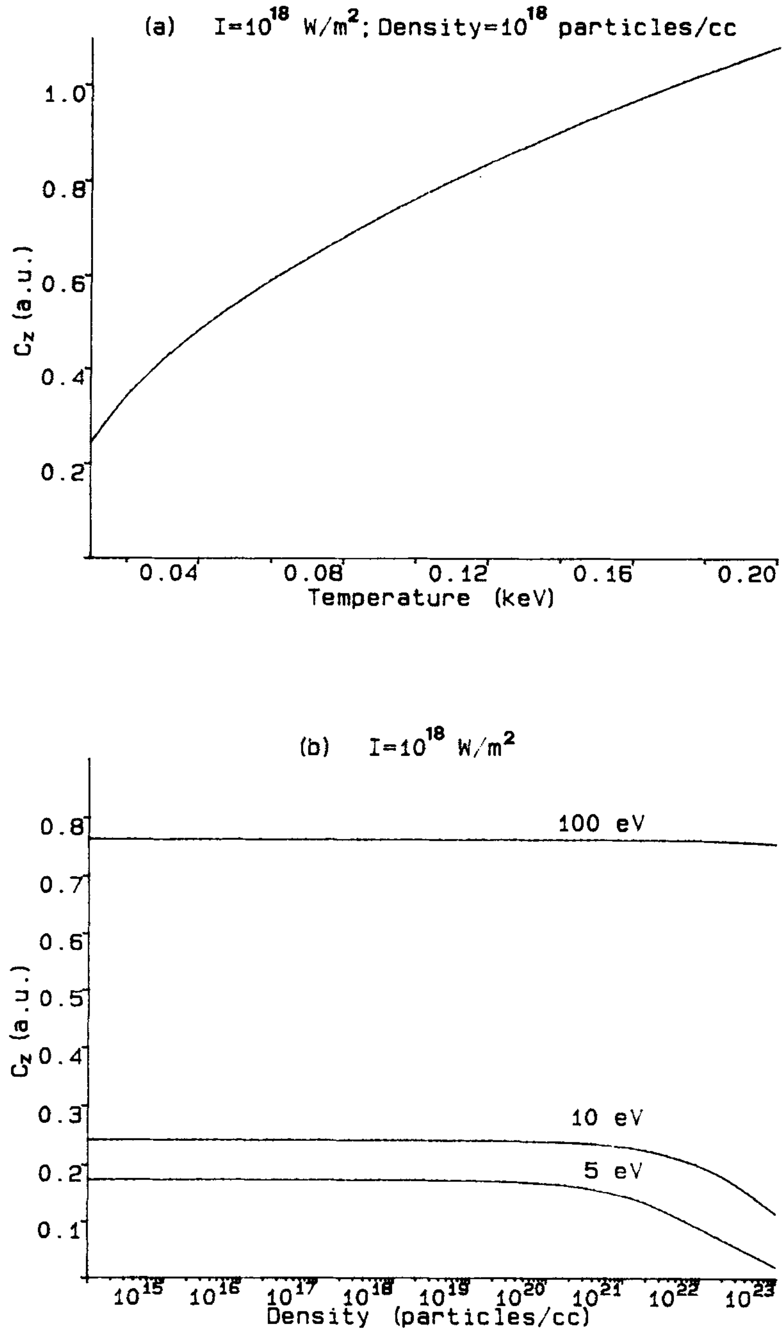

Figure 1 Variation of non-degenerate density C_z in Eq. (2.7) with: (a) temperature at fixed density 10^{18} particles/cc; (b) density at the temperatures corresponding to $k_BT = 5$, 10 and 100 eV. In the calculation $z = z_0$ and $z \ll E/\omega^2$. The equivalent laser flux is 10^{18} Watt/m^2.

considered, this then implying the constraint that the ionic number density n_i satisfies

$$n_i \ll 1.4 \cdot 10^{23} \frac{1}{Z} \left(\frac{k_B T}{10eV}\right)^{3/2} \tag{4.1}$$

with Ze the charge of an ion. This delineates the region of classical plasmas. The most recent experiments have considered higher densities, where the effect of the degeneracy of the electrons becomes important. However in the context of multi-photon ionization relatively low density plasmas are normally investigated. The presently achievable laser flux is about 10^{18} Watt/m^2. The connection between the laser flux and the electric field is given by

$$I = cE_{\max}^2/8\pi. \tag{4.2}$$

In the above calculation we assumed a static electric field. Brewczyk and Gajda[3] pointed out under what conditions this is a reasonable assumption.

Figure 1 shows the temperature, density and electric field dependence as represented by Eq. (2.7). The behaviour of C_z in the density and temperature region on which we focus is dominated by the temperature dependence which is $\sim T^{1/2}$. The E-field and density dependence is the stronger the lower the temperature. The plot is for $z = z_0$ and z substantially less than E/ω^2. Figure 2 similarly shows how the x

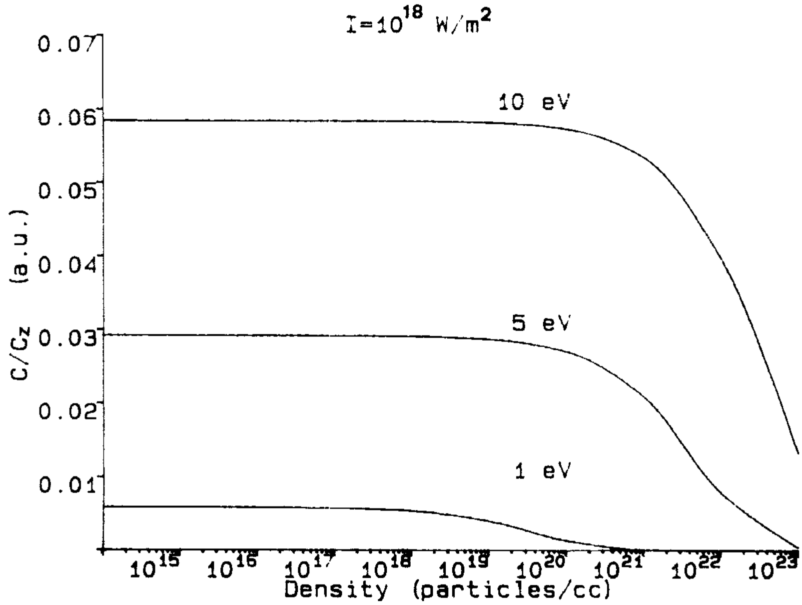

Figure 2 Same as Figure 1(b) but now for the x and y contributions to the non-degenerate density C from Eq. (2.9), and with different temperatures corresponding to $k_B T = 1$, 5 and 10 eV. It should be noted that for $k_B T = 1$ eV, C begins to decrease at densities of $\sim 10^{18}$–10^{19} particles/cc.

and y contribute to C, namely how the ratio C/C_z, from Eq. (2.9), depends on temperature and density. Here the temperature dependence is $\sim T$ and again the density dependence is the stronger the lower the temperature.

5 SUMMARY AND FUTURE DIRECTIONS

In this paper, closed forms have been obtained for the canonical density matrix C for electrons moving in a static electric field E, and confined by a harmonic restoring force. Model potentials $V(\mathbf{r})$ have then been 'switched on' to this above canonical density matrix via the TF approximation (3.1).

It would be of interest to apply the method of March and Murray[9] to convert C, the electron density for non-degenerate electrons, into results applicable to intermediate degeneracy governed by Fermi–Dirac statistics. Unfortunately, without switching on the model potential $V(\mathbf{r})$, this is already difficult to handle by purely analytical methods, as can be seen from the case of complete degeneracy for the harmonic oscillator alone in Refs [6] and [7]. No doubt, numerical procedures will eventually enable our present results to be transformed according to the route established in Ref. [9].

The same situation obtains when one attempts to remove the TF approximation underlying Eq. (3.1). With $C^{(0)}$ instead of the free-particle C_0, the generalization of the Green function G_0 in Eq. (3.3) is hard to effect analytically. Numerical presentation will be difficult, because of the large number of variables involved.

Nevertheless, it seems to us likely that the model treatment of atomic ions in hot, non-degenerate plasmas presented in this work, is well worth further study, the intermediate Fermi–Dirac degeneracy being of obvious importance. Under these conditions, an appropriate starting point to introduce the potential would be the elevated temperature Thomas–Fermi theory[10].

Acknowledgements

One of us (C.A.) wishes to acknowledge the award of a NATO-CNR Fellowship which supported his stay in Oxford. Another of us (S.P.) acknowledges financial support from the University of Belfast, during the period in which her contribution to the present study was made. She wishes also to thank Dr. S. Rose of the Rutherford Laboratory for valuable discussions on this general area and for his continuing interest in this work.

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APPENDIX

The approach of Hilton *et al.*[8] can be generalized for any reference Hamiltonian for which the solution of the relevant Bloch equation is known. For a given Hamiltonian

$$\hat{H}_0 = -\tfrac{1}{2}\nabla^2 + V_0(\mathbf{r}) \tag{A.1}$$

it is possible to find the solution of the Bloch equation for a perturbed Hamiltonian

$$\hat{H} = \hat{H}_0 + V(\mathbf{r}). \tag{A.2}$$

The procedure is to write

$$C(\mathbf{r}, \mathbf{r}_0, \beta) = C_0(\mathbf{r}, \mathbf{r}_0, \beta) \exp[-\beta U(\mathbf{r}, \mathbf{r}_0, \beta)] \tag{A.3}$$

where $C_0(\mathbf{r}, \mathbf{r}_0, \beta)$ satisfies the equation

$$\hat{H}_0 C_0 = -\frac{\partial C_0}{\partial \beta}. \tag{A.4}$$

The effective potential matrix $U(\mathbf{r}, \mathbf{r}_0, \beta)$ satisfies the equation

$$\left[1 + \frac{\partial}{\partial \beta} - \beta \frac{\vec{\nabla} C_0}{C_0} \cdot \vec{\nabla} - \frac{\beta}{2}\nabla^2\right] U = V - \tfrac{1}{2}\beta^2 |\vec{\nabla} U|^2. \tag{A.5}$$

As in the approach of Hilton *et al* this differential equation in U can be transformed into an integral equation by using the Green function of the left-hand side operator in Eq. (A.5). This Green function maintains the same form as for the solution of Hilton *et al* for the perturbed non-interacting free-electron system, namely

$$G(\mathbf{r}, \mathbf{r}_0, \mathbf{r}_1, \beta, \beta_1) = \frac{C_0(\mathbf{r}, \mathbf{r}_1, \beta - \beta_1) C_0(\mathbf{r}_1, \mathbf{r}_0, \beta_1)}{\beta C_0(\mathbf{r}, \mathbf{r}_0, \beta)} \theta(\beta - \beta_1) \tag{A.6}$$

but now C_0 is the solution of the Bloch Eq. (A.4) for the reference Hamiltonian (A.1). The corresponding integral Eq. to (A.5) is then

$$U(\mathbf{r}, \mathbf{r}_0, \beta) = \int d\mathbf{r}_1 \int_0^\beta d\beta_1 G(\mathbf{r}, \mathbf{r}_0, \mathbf{r}_1, \beta, \beta_1)\left[V(\mathbf{r}_1) - \frac{\beta^2}{2}|\vec{\nabla} U|^2\right] \tag{A.7}$$

When it is possible to neglect the term $(\beta^2/2)|\vec{\nabla} U|^2$, Eq. (A.7) gives a direct route for calculating the "effective potential" U. This linear response treatment can be applied under the following conditions: (i) U small and much more slowly varying with $\mathbf{r}$ than $V(\mathbf{r})$, especially in presence of Coulomb singularities, (ii) $|\vec{\nabla} U|^2$ small, (iii) β small. Using Eq. (2.9) for C_0 in (A.6) and (A.7) it is not possible to give an analytical expression for U even in the linear response approximation and numerical procedures are required. When it is possible to make the assumption $\beta\omega < 0.5$ the β-convolution in Eq. (A.7) can be computed by approximating the hyperbolic functions of C_0 by the lowest order powers in $\beta\omega$. In the linear response approximation in V and in this high temperature regime, Eq. (A.7) takes the form, for the diagonal elements,

$$U(\mathbf{r}, \beta) = \int d\mathbf{r}_1 V(\mathbf{r}_1) G(\mathbf{r}, \mathbf{r}_1, \beta) \tag{A.8}$$

where

$$G(\mathbf{r}, \mathbf{r}_1, \beta) = \frac{1}{\pi\beta|\mathbf{r}_1 - \mathbf{r}|} \exp\left\{ -\frac{2(\mathbf{r}_1 - \mathbf{r})^2}{\beta} + \frac{\beta E}{2}(z_1 - z) - \frac{\beta\omega^2}{8}[(\mathbf{r}_1 + \mathbf{r})^2 - 4r^2] - \frac{\beta\omega^2}{24}(\mathbf{r}_1 - \mathbf{r})^2 \right\}. \tag{A.9}$$

This reduces to the free-electron Green function when $E = 0$ and $\omega = 0$.