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On a Dielectric Formulation of the Theory of Electro-Migration in Metals

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Abstract. A simple and approximate theory of electro-migration in metals is formulated within the dielectric function approach by the use of the 'equivalence theorem' discussed earlier by Peierls and the author. In the case of an interstitial ion of weak charge, the result of earlier work by several authors follows from this formulation. The scope and the limitations of the method are discussed. The case of an ion electro-migrating in a charged Bose gas is commented on. Finally the effect of ion-ion interaction on the force in electro-migration is considered.

1. Introduction

In this paper we shall deal with the theory of a deceptively simple phenomenon in metals and a problem of long standing. It is the problem of electro-migration – the migration of ionic impurities in the presence of an external electric field usually setting up a steady state current in metals. In recent years this problem has drawn considerable interest and has been looked at from various points of view. The theories of Fiks [1] and of Huntington and Grone [2] treat the phenomenon as essentially a competition between the direct field force and that due to the so-called 'electron wind' arising from the momentum transfer by the streaming electrons to the ion. Bosvieux and Friedel [3] calculate the electron wind term from the electron density redistribution around the moving ion. This is done through a first order perturbation, the ionic charge being assumed as weak. The work of Sorbello [4] is also along similar lines. Sorbello's result seems to be sensitive to the choice of the pseudo-potential which is used in his theory to calculate the perturbed electron density.

These theories e.g. those of Fiks and of Huntington and Grone either do not deal with the tricky question of screening around the moving ion and its possible effect on electro-migration, or assume (e.g. Bosvieux and Friedel) that an interstitially migrating ion is completely screened off from the external electric field, the only other force exerted on it being the electron wind force. Sorbello acknowledges that the situation regarding screening is not quite clear.

Peierls and co-workers have tried to understand the question of screening in a simple semi-classical model and have come to some interesting conclusions with regard to any possible contribution to the force from the screening. We refer to the work of Das and Peierls [5] for an account of their findings.

Recently Peierls and the author [6] have shown, how, in their model, the force on the ion can be expressed exactly (exact in the ionic charge) in terms of the resistivity

change due to the ion. The force also contains the contribution from the so-called 'carrier density modulation' to which Landauer [7] has drawn attention. We shall come back to this point in Section 3.

We shall explore here a dielectric formulation of the theory of electro-migration in simple metals. We believe that such a formulation constitutes a useful step toward a more satisfactory microscopic theory. The latter is regarded as rather difficult. We find that by exploiting the 'equivalence theorem' discussed by Das and Peierls [5], a formulation in terms of the dielectric function can be given. In the case of an ion of weak charge (the linear response to the ionic charge) migrating interstitially, the result of Bosvieux and Friedel (BF) follows from this formulation. A little rearrangement leads also to Sorbello's result for the same case. The method is described in the next section. The final section contains a discussion of the method and its possible extension.

2. Dielectric Formulation

The idea behind our formulation is as follows: one obtains a self-consistent velocity-dependent potential for an ion of charge Ze (treated as weak) moving slowly with velocity \mathbf{u} ($|\mathbf{u}| \ll v_F$, the Fermi velocity of the electrons) in a metal in a steady state in an external electric field E . The metal is approximated by the familiar 'jellium' model. The electron-electron interaction is considered in the Hartree approximation. The exchange and the correlation conserve momentum and hence would not affect our conclusions in any qualitative manner. The lattice is practically ignored except perhaps for the effective mass of the electrons. Some phonon mechanism could be introduced. This could be done in the frame-work of the jellium model in order to avoid the complications of a crystal lattice. The electrons in the steady state are assumed to have a constant collision time τ . This allows us (see below) to utilize the 'equivalence theorem'. We consider the metal at very low temperatures $T \simeq 0^\circ\text{K}$. But the temperature correction can be calculated.

In the present formulation it is important to work in the appropriate frame of reference, and it seems worthwhile to elaborate on it.

We choose a frame of reference in which the ion of charge Ze is at rest, and the jellium background is moving with a relative velocity $-\mathbf{u}$. However it is not obvious how the external electric field will enter the problem. For this purpose we invoke the 'equivalence theorem' which states that for a constant collision time τ we can replace $(eE/m)\tau$ in favour of \mathbf{u} with equivalent results. We have detailed the argument in Appendix A. We have decided to present it in an appendix because the 'equivalence theorem' originated from a semi-classical transport theory whose formal set-up is somewhat different from that of this paper. We now recognize that in our frame of reference, the electrons having velocity $\mathbf{u} \Leftrightarrow (eE/m)\tau$, τ being finite, are responding to a stationary ion. It is necessary that this τ remains finite; otherwise a steady state will not obtain. But after the identification between \mathbf{u} and E has been made, we regard the electrons (or the quasi-particles on the Fermi surface) responding to the stationary ionic charge alone, as having an infinite life time. We should however add that the last assumption is not necessary in the general theory of linear response to the moving charge. But in this note we have decided to confine ourselves to the linear response theory derived along traditional lines (see e.g. Lindhard [8]) in which the finiteness of the relaxation time is disregarded. We shall take up this point in Section 3. In our present calculation we shall consider the traditional linear response theory in the random phase approximation.

The velocity-dependent potential $V(\mathbf{q}, \omega)$ of the ion can be written as

$$V(\mathbf{q}, \omega) = V_0(\mathbf{q})/\epsilon(\mathbf{q}, \omega) \tag{1}$$

where $V_0(\mathbf{q}) = -4\pi Ze/q^2$ and $\epsilon(\mathbf{q}, \omega)$, the RPA dielectric function is

$$\begin{aligned} \epsilon(\mathbf{q}, \omega) &= 1 - \frac{4\pi e}{q^2} \lim_{s \rightarrow 0^+} \sum_{\mathbf{k}} \frac{f_0(E_{\mathbf{k}+\mathbf{q}}) - f_0(E_{\mathbf{k}})}{E_{\mathbf{k}+\mathbf{q}} - E_{\mathbf{k}} - \omega + is} \\ &\equiv 1 - \frac{X(\mathbf{q}, \omega)}{q^2}, \quad (\text{we have put } \hbar = 1). \end{aligned} \tag{2}$$

The external electric field will now be introduced in the theory through the drift velocity \mathbf{u} , given by $\omega = \mathbf{q} \cdot \mathbf{u}$. Assuming the electric field to be in z -direction, we can write $\omega = q_z u$. u will then be replaced by E according to the ‘equivalence theorem’. We have assumed $u \ll v_F$, and hence we shall calculate $V(\mathbf{q}, u)$ only up to the order (u/v_F) . It can be seen that this corresponds to the Ohmic region of conductivity.

Let us write

$$V(\mathbf{q}, u) = V^{(0)}(\mathbf{q}, 0) + V^{(1)}(\mathbf{q}, u) \tag{3}$$

where $V^{(1)} \propto (u/v_F)$ the evaluation of $X(\mathbf{q}, u)$ is too familiar to be detailed here. We write

$$X(\mathbf{q}, u) = X_1(\mathbf{q}, u) + iX_2(\mathbf{q}, u) \tag{4}$$

$$\begin{aligned} X_1(\mathbf{q}, u) &= \frac{\alpha^2}{2} \left[1 + \frac{4k_F^2 - (q - 2\lambda)^2}{8k_F q} \ln \left| \frac{2k_F + q - 2\lambda}{2k_F - q + 2\lambda} \right| \right. \\ &\quad \left. + \frac{4k_F^2 - (q + 2\lambda)^2}{8k_F q} \ln \left| \frac{2k_F + q + 2\lambda}{2k_F - q - 2\lambda} \right| \right] \end{aligned} \tag{5}$$

where $\lambda = muq_z/q$ and $\alpha = (\text{Thomas-Fermi screening length})^{-1} = [32\pi^2 e^2 m k_F / (2\pi)^3]^{1/2}$.

It is easy to see that $X_1(\mathbf{q}, u)$ is even in u . This follows from the dispersion relations as well.

$$X_1(\mathbf{q}, u) \approx X_1(\mathbf{q}, 0) + 0(u^2). \tag{6}$$

We next consider $X_2(\mathbf{q}, u)$. Again the calculation is standard. For the value of u of our interest

$$X_2(\mathbf{q}, u) = -\frac{\pi}{2} \alpha^2 \frac{uq_z}{v_F q}. \tag{7}$$

We now go back to equation (1) and recall equations (6) and (7).

$$\begin{aligned} V(\mathbf{q}, u) &= V_0(\mathbf{q}) \left[1 - \frac{X(\mathbf{q}, u)}{q^2} \right]^{-1} \\ &\approx V_0(\mathbf{q}) \left[1 - \frac{1}{q^2} \left\{ X_1(\mathbf{q}, 0) - \frac{i\pi}{2} \alpha^2 \frac{uq_z}{v_F q} + 0(u^2) \right\} \right]^{-1}. \end{aligned}$$

A little algebra leads to

$$V(\mathbf{q}, u) \approx \frac{V_0(\mathbf{q})}{[1 - X_1(\mathbf{q}, 0)/q^2]} - i\pi \frac{\alpha^2 q_z}{q^3} \frac{u}{v_F} \frac{V_0(\mathbf{q})}{[1 - X_1(\mathbf{q}, 0)/q^2]^2}. \tag{8}$$

Let us analyze equation (8). The first term is $V^{(0)}(\mathbf{q}, 0)$. We see that in zeroth order in u , the screening cloud is isotropically distributed around the ion. Note that the long-range Friedel–Kohn oscillations are still present; but these are isotropic.

The second term in (8) is $V^{(1)}(\mathbf{q}, u)$, the velocity-dependent potential due to the slowly moving ion. This term indicates that the screening cloud is distributed anisotropically around the moving ion—it is, so to say, elongated along the direction of the electric field.¹⁾ This term has an interesting feature: if one takes the limit $\tau \rightarrow \infty$ in the u -dependent potential derived in Das and Peierls [5] for small Ze (equations (4.16) and (4.21) of their paper), it reduces to the second term of (8). This comparison is instructive and has some conceptual content. This will be further commented on in the next section.

In the limit of an infinitely heavy ion, we can write the force on the ion of charge Ze , as

$$F_z(0) = Ze \left. \frac{\partial V(\mathbf{r}, u)}{\partial z} \right]_{r \rightarrow 0} \quad (9)$$

where

$$V(\mathbf{r}, u) = \frac{1}{(2\pi)^3} \int e^{-\mathbf{q} \cdot \mathbf{r}} V(\mathbf{q}, u) d^3\mathbf{q}. \quad (10)$$

Therefore

$$F_z(0) = \frac{iZe}{(2\pi)^3} \int q_z V(\mathbf{q}, u) d^3\mathbf{q}. \quad (11)$$

It is clear that the first term in (8) will not contribute to the force. Considering the second term

$$F_z(0) = \frac{\pi \alpha^2 Z^2 e^2}{2} \frac{u}{2\pi^2} \frac{1}{v_F} \int \frac{q_z^2 d^3\mathbf{q}}{q^5 [1 - X_1(\mathbf{q}, 0)/q^2]^2}. \quad (12)$$

This force, in our formalism, is in addition to the direct field force $Ze\mathbf{E}$ on the ion. The latter is derivable from the asymptotic value

$$Ze \left. \frac{\partial V(\mathbf{r}, u)}{\partial z} \right]_{r \rightarrow \infty}.$$

We would like to say more about this term in the next section. We can now replace \mathbf{u} in (12) by $(e\mathbf{E}/m)\tau$ ('equivalence theorem'). The physical interpretation of (12) is then that it is the electron wind force. As anticipated, this term is independent of the sign of the ionic charge but depends on that of the carrier charge – the direction will be opposite for the hole carriers. In fact it is easy to see that (12) is the same force on an interstitial ion calculated by BF (equation (III.15) of their paper). In order to recover the result of Sorbello (equation (4.11) of his paper) one needs to disentangle the dielectric function from the pseudo-potential used by Sorbello. When the latter is taken as a Coulombic potential, Sorbello's result is the same as (12)²⁾.

¹⁾ It should be pointed out that the asymptotic dipolar behaviour (at distances $> 1/k_F$) of the induced electron density is also present. Actually the electron distribution asymptotically is a complicated superposition of the dipolar and the Friedel–Kohn oscillatory terms.

²⁾ It is possible that one can arrive at (12) by a Hellman–Feynman type argument (see for example Slater [9]), in place of (9). However, in our view, the application of Hellman–Feynman type argument to a problem of steady state requires a careful analysis, and we are not aware of any general proof.

According to BF, (12) is the only force on the migrating ion. For conclusions to the contrary, see Das and Peierls [5, 6].

The dielectric formulation can also be extended, with reasonable validity, to surface electro-migration in metals. One can use the dielectric function derived e.g. by Newns [10] for a bounded jellium. Extra complications would arise since the response matrix will no longer be diagonal. It is now well established that the screening of an ion on a metal surface is qualitatively different from that in the bulk [10]. There is also the question of the gravitationally induced electric field near a metallic surface. (See Schiff [11] and other references therein.) The study of this electric field has generated considerable interest, though the quantitative estimates have remained controversial.) Such an electric field should make additional contribution to the force on the ion. Surface electro-migration is expected to show some new features. We believe this problem merits a separate study.

The result (12) is more applicable to the case of an interstitially migrating ion because the lattice distortion has been neglected in our formalism.

Discussion

Let us now make some observations on the dielectric function approach as outlined here. The approach looks attractive and has certain unifying properties. It is however useful to recognize the assumptions and the approximations of the formulation. The external electric field has been introduced in the theory through the use of the 'equivalence theorem' which relies on the assumption of a constant collision time. For simple (non-transitional) metals this is a reasonably good assumption. As we have observed in Appendix A, in the case of an energy-dependent collision time (viz. for a transition metal) the 'equivalence theorem' would break down. In such cases it may not be easy to give a dielectric formulation of electro-migration.

We have noted, parenthetically, that the result of BF and of Sorbello for the electron wind term follows from our method in an approximation that is tantamount to an infinite relaxation time dielectric formulation. It should be stated that such a formulation is not quite satisfactory because the problem of electro-migration concerns a steady state characterized by a finite relaxation time. The arguments of Section 1 are given validity by the use of the moving frame and the 'equivalence theorem'. Another aspect of the infinite relaxation time approximation has some interesting physical implication. It is well known that such a formulation violates the continuity equation or in other words the local charge conservation. As remarked in Section 1, one can relax this restriction. A fully microscopic calculation of the dielectric function for a finite relaxation time is a difficult task. A simple approach has recently been discussed by the author [12], which utilizes a 'local equilibrium distribution' and hence a 'local chemical potential' for the electrons³⁾. Using the dielectric function derived there, we

³⁾ We are considering a degenerate electron gas. If we consider a charged Bose gas (CBG), that may undergo a Bose-Einstein condensation, the local chemical potential would tend to vanish, thereby inhibiting the local charge conservation. It has been argued by the author [13] that the screening in a CBG should be significantly affected in case of condensation. Two recent calculations [14, 15] bear this out. It then follows that the force on an electromigrating ion in a CBG should be affected correspondingly. A CBG has still remained as a curiosity of the theoretical physicists, though in astrophysical objects such as a neutron star charged Bose matter is believed to exist. But to date it has not been realized in a laboratory. Here we make a suggestion: it is now experimentally feasible to inject deuterium at very high density into some metals. It will be interesting to search for some properties of a CBG e.g. Meissner effect [16], in these systems.

can, again in the linear Ze approximation, calculate the force on the ion. It is found that the force (apart from the direct field term) will now have an extra term. We interpret this term as a contribution from the ‘carrier density modulation’ (CDM) to order $(Ze)^2$. In order to appreciate it, let us briefly recall, in a simplified version, the arguments of Das and Peierls [6] on the contribution from the CDM to the force. As Landauer [7] has pointed out, the ion will cause both localized scattering of the carriers as well as a spatial modulation in carrier density. One can perhaps coin the phrase ‘convective contribution to transport coefficients’ for the second process.

In a real three dimensional metal it is hard to separate these two effects. A semi-classical one dimensional case can perhaps better illustrate the latter effect (which is the CDM), because in this case the ionic potential causes no scattering. It is therefore the CDM which will result in a spatial variation of the field around the ion in the steady state.

Peierls and the author have derived an exact (exact in the ionic charge) expression for the total force in terms of the integral of the current and hence in terms of the resistivity change due to the ion. As we have just remarked, it is not easy to separate out the CDM contribution to this additional resistivity. In the model adopted by Das and Peierls, the local change in the conductivity is proportional to the change in the local carrier density. (This is also the case in our dielectric formulation.) Peierls and the author have argued that in the case of small Ze i.e. for weak CDM, the spatial variation of the field can be neglected up to a correction of order $(Ze)^2$, and the direct field term in the force can be restored through the CDM contribution to the resistivity change, and invoking the local charge conservation. This correction from the CDM arises in addition to the electron wind term in the same order.

From the above considerations we see the role of the local charge conservation in the dielectric formulation of electro-migration: some vestiges of the CDM enter the force through this requirement.

The exact theory of Peierls and the author thus contains all the contributions to the force, including the direct field term. When we visualize the exact expression for the force as an expansion in powers of Ze , the second order terms make contact with those obtained through the dielectric formulation. Since in the dielectric formulation as outlined here a small Ze is assumed from the beginning, the local inhomogeneity of the field could be neglected, and the direct field term can be obtained asymptotically, as remarked in Section 2. But it would not be so straightforward in a non-linear dielectric formulation. The linear response theory of this note seems to be a useful first step toward such more exact microscopic theory.

We conclude with a remark on another aspect of the problem that has not yet received sufficient active consideration in the literature. We have so far assumed a dilute distribution of migrating ions so that they may not interact among themselves. Such an arrangement is of course hard to realize in practice. It is therefore useful to know what new complications may arise in the case of interacting ions. The simplest case will be to consider only two ions interacting through the electron gas. This can be studied through the pseudopotential scheme [17]. But the method relies on the ionic potential being assumed weak. Though this will be consistent with the spirit of our dielectric formulation, we have described in Appendix B, a simple diagrammatic technique which is not restricted to this assumption. We believe this method can be of use in similar problems in other areas. The extension of the method to multi-ion interaction does not look straightforward. The technique to calculate three and four-body forces etc. in nuclear matter might be a useful guide in this regard.

We have shown in Appendix B that when one includes a screened ion-ion interaction, an extra term will appear in the force. This term depends on the sign of the ionic charge and cannot be interpreted as an electron wind force. In fact the interpretation of this term and others that will result from higher order ion-ion interaction does not look simple. The calculation in the Appendix should therefore serve to illustrate the additional conceptual difficulty in the theory of electro-migration allowing for a more realistic situation in metals.

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Appendix A

The ‘equivalence theorem’

In this appendix we reproduce the essence of the ‘equivalence theorem’ as discussed previously by Peierls and the author [5].

Let us recall the semi-classical Boltzmann equation approach of Ref. [5]. In the ‘co-moving frame’ the equation is

$$\frac{\mathbf{p}}{m} \cdot \frac{\partial n_1}{\partial \mathbf{r}} - \frac{\partial V_0}{\partial \mathbf{r}} \cdot \frac{\partial n_1}{\partial \mathbf{p}} - \frac{1}{\tau} (\bar{n}_1 - n_1) = \left(\frac{1}{m} \frac{\partial V_1}{\partial \mathbf{r}} - \frac{\mathbf{u}}{\tau} \right) \cdot \mathbf{p} \frac{\partial n_0}{\partial \epsilon}. \tag{A.1}$$

In equation (A.1), n_1 is the first order (proportional to the electric field E and the ionic velocity u) deviation from the equilibrium distribution n_0 for the electrons. V_0 is the potential for the stationary ion and V_1 is the same for the moving ion. V_0 and V_1 are to be determined from the Poisson equations which need not concern us here. The detailed derivations of these equations are in Ref. [5].

The electric field is given by

$$V_1 \rightarrow eEz, \quad r \rightarrow \infty. \tag{A.2}$$

We now observe that equation (A.1) remains unchanged if we replace u by

$$u' = u + v \tag{A.3}$$

and V_1 by

$$V_1'(\mathbf{r}) = V_1(\mathbf{r}) + \frac{mv}{\tau} z \tag{A.4}$$

which by equation (A.3) means that the external field is replaced by

$$E' = E + \frac{mv}{e\tau}. \tag{A.5}$$

This is the ‘equivalence theorem’. This indicates that the effect on the electron distribution of an electric field and of the motion of the ion is the same. The physical origin of this identity is as follows: the effect of the field on the electrons of the pure metal (far from the ion) is to give them a uniform drift velocity, superimposed on their normal motion. If the ion moves with the same velocity, then the relative velocities

remain the same. Therefore all results of interaction such as the electron density distribution remain the same.

We see that the assumption of a constant collision time plays an important role in arriving at the ‘equivalence theorem’. If different groups of electrons have different collision times, they would have different drift velocities for a given field, and hence we could not have an ‘equivalence theorem’. This would be the case e.g. for a two-band metal consisting of both electron and hole carriers.

The ‘equivalence theorem’ should remain valid in the presence of a crystal lattice. Because we can still write the transport equation in the ‘co-moving frame’, and the equations (A.3)–(A.5) of this appendix should still hold.

Appendix B

Electro-migration of interacting ions

Let us consider two ions, each of charge Ze , moving with the same velocity u in the jellium. Therefore by the arguments of Section 1, we can assume the ions at rest, and the jellium now in the moving frame. (In reality the two ions may not be moving with the same velocity.)

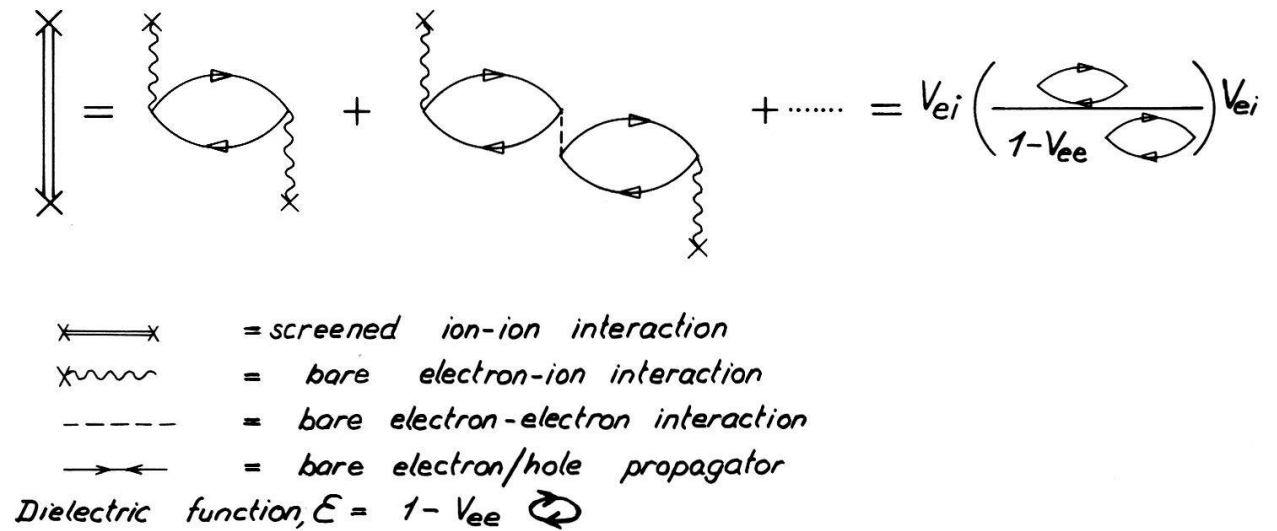


Figure 1

Figure 1 summarizes the diagrammatic method to find the screened ion-ion potential $V_{ii}(\mathbf{q}, \omega)$. The result is

$$V_{ii}(\mathbf{q}, \omega) = \frac{[V_{ei}(\mathbf{q})]^2}{[V_{ee}]} \left[\frac{1}{\epsilon(\mathbf{q}, \omega)} - 1 \right] \tag{B.1}$$

where

$$\begin{aligned} V_{ei}(\mathbf{q}) &= \text{electron-ion potential} = -4\pi Ze/q^2, \text{ and} \\ V_{ee}(\mathbf{q}) &= \text{electron-electron potential} = 4\pi e/q^2. \end{aligned} \tag{B.2}$$

The potential $V_{ii}(\mathbf{q}, \omega)$ can be added to the screened ionic potential $V(\mathbf{q}, \omega)$ obtained in Section 2, to get the total potential $V_T(\mathbf{q}, \omega)$. If we approximate $V_T(\mathbf{q}, \omega)$ according to the procedure of Section 2, we find

$$V_T(\mathbf{q}, u) \approx V_T^{(0)}(\mathbf{q}, 0) + V^{(1)}(\mathbf{q}, u) + V_{\text{ion-ion}}(\mathbf{q}, u) \tag{B.3}$$

with

$$V^{(1)}(\mathbf{q}, u) = -\frac{i\pi}{2} \alpha^2 \frac{q_z}{q} \frac{u}{v_F} \frac{V_0(\mathbf{q})}{[1 - X_1(\mathbf{q}, 0)/q^2]^2} \quad (\text{B.4})$$

and

$$V_{\text{ion-ion}}(\mathbf{q}, u) = -\frac{i\pi}{2} \alpha^2 \frac{q_z}{q} \frac{u}{v_F} \frac{[V_0(\mathbf{q})]^2}{[V_{ee}]} \frac{1}{[1 - X_1(\mathbf{q}, 0)/q^2]^2}. \quad (\text{B.5})$$

In $V_{\text{ion-ion}}$ we have retained only that term which will give a non-zero contribution to the force. The force $F_z(0)$ on the ion will now have an extra term, which looks structurally the same as $V^{(1)}(\mathbf{q}, u)$ but with a different coefficient and with one difference: it is proportional to $(Ze)^3 E\tau$, not a characteristic of the wind force.

We may add that the diagrammatic method can accommodate $\epsilon(\mathbf{q}, \omega)$ beyond the time-dependent Hartree approximation of Section 2. The electrons can also be assigned any energy band structure. The method can be applied to surface electro-migration as well.

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