Geometry and boundary conditions in the Das-Peierls electromigration theorem

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In a set of closely related papers, one by Das and Peierls, the other by this author, identical expressions were derived for the driving force in electromigration. Two later papers, one by Sham, the other by Turban, Nozières and Gerl, cite the same expression. It is shown that the apparent agreement by Turban, Nozières, and Gerl is not a result of real agreement about the detailed physics of the process. The argument given by Sham has some ambiguity about geometry and boundary conditions, but is closer to that of the two earlier papers.

In a set of closely related papers, one by Das and Peierls,[1] the other by this author,[2] identical expressions were derived for the driving force in electromigration. This author has called this result the Das-Peierls theorem. For a set of lattice defects in "jellium," the force per unit volume given by the Das-Peierls theorem is

$$-E_T e n_0(\rho - \rho_0)/\rho \ .$$

Here E_T is the space-average field in the material containing the defects and ρ is the resistivity of that material. ρ_0 and n_0 are the resistivity and carrier density in the pure material, without the defects. Two other papers, one by Sham,[3] the other by Turban, Nozières, and Gerl[4] use momentum-balance arguments to derive the same expression. Indeed two further recent notes[5,6] point to the agreement between the four results.[1-4] Here we will discuss whether the apparent exact agreement of the momentum-balance considerations in Refs. 3 and 4 with those of Refs. 1 and 2 is a result of real agreement about the detailed physics of the process. Even without attention to the details, it is clear that Refs. 1 and 2 exhibit a good deal of concern for boundary conditions, not found in Refs. 3 and 4. Ref. 2 introduced a particular geometrical configuration which at first sight may seem a little complex to the reader. It was actually introduced, however, to emphasize the difference between the space-average field in the sample, and that portion of the field arising from charges outside the sample (or at its very edge). That distinction, however, is not important for very dilute alloys, with a very small residual resistivity. The derivation in Ref. 2 is *not* limited to the dilute case.

We will, in this paper, focus on electromigration in a slab of limited thickness, with plane-parallel electrodes. Unfortunately, Refs. 3 and 4 do not mention geometry and boundary conditions. An alternative geometry to the slab is that of a torus. The use of a torus to deduce the driving force for electromigration from momentum balance argument has been invoked in the past.[7,8] A torus is not without complications of its own. Contrary to the prevailing assumption among theoreticians a torus is not a slab with periodic boundary conditions. The torus has surfaces which complicate electrostatic considerations. It is now generally agreed that current flow in the presence of lattice defects is accompanied by nonuniform fields and that the electronic charge distribution near the defect is polarized. In the case of a torus such a local polarization must be accompanied by charges on the surface of a torus which guide the lines of force, emanating from the local polarization, around the circular shape of the torus. While neither Sham[3] nor Turban *et al.*[4] specify a torus, Sham, in his Eq. (2.2), uses a time-dependent vector potential as the source of the electric field. This is at least somewhat suggestive of the toroidal geometry. By contrast, Turban *et al.*, in their Eq. (19), as well as by their references to electrons, "...free to reach the end of the crystal," clearly invoke a terminated linear sample. In any case, we will leave it to others to do a careful electrostatic analysis of the torus, and we will only discuss the terminated sample. Sham's momentum-balance equations, as given in Sec. III E of Ref. 3 are, in fact, likely to be directly applicable to the case of a torus. Since, however, I have not carried out the required detailed electrostatic analysis of the torus I will not make that as a formal claim. The analysis of Turban *et al.* is also likely to be more applicable to a torus, as indicated by Gerl[9] and Nozières.[10] In their case, however, not only is that more clearly in contradiction to the actual content of their paper, it is also clearly inadequate as a "repair" for all of the subsequent deficiencies to be discussed.

Let us first discuss Sham's analysis. We limit our considerations to one short section in Sham's paper, Sec. III E. This was clearly intended to be a simplification of Refs. 1 and 2, (of this

present paper) as a service to the reader. Nevertheless we can ask whether it is a valid simplification. As already emphasized, we shall consider a slab geometry, even though that may not be the best way to view Sham's equations. Sham considers the forces on all the conduction electrons, and sets this sum equal to zero. One of the terms represents the force of the bare defect ions on the electron gas, and this is the negative of the driving force exerted by the electron gas polarization on the ions. So far Sham's procedure agrees with that in Refs. 1 and 2. Sham in his force balance, however, does not include the interaction between the polarized electron gas and the uniform ionic charge of the original unperturbed lattice.

To make this point in more detail, we must consider boundary conditions. If an electronic polarization is established near a defect, the resulting field is determined not only by the charge distribution, but also by the boundary conditions. For a sensible discussion of boundary conditons we must specify a particular physical environment for the sample. Let us assume that the specimen is terminated by highly conducting equipotential electrodes. Two obvious boundary conditions: (i) Maintain the total voltage across the sample as the polarization is established. (ii) Maintain the integrated flux of the field across far away electrodes as the polarization is established. Let us assume that a boundary layer, near each electrode, roughly a few mean free paths in thickness, contains no impurity atoms. Then it becomes clear that this boundary condition (BC) corresponds to maintaining the original current flow. At the same time the thin layer of pure metal will cause a negligible change in the average properties of the sample, as well as in the environment (and therefore in the electromigration force) seen by most of the impurities. (The pure boundary layer is a tutorial simplification, and not an essential part of our argument.)

The effect of these boundary conditions is most easily understood by superposing a uniform distribution of similar polarization patterns, in which the polarization charges are displaced transverse to the direction of the dipole moment. We thus simplify to an essentially one-dimensional problem. As long as we are concerned only with quantities integrated over planes perpendicular to the direction of current flow it does not matter whether we consider the electrostatics of three-dimensionally localized dipoles, or of dipole sheets.

Let us discuss the BC (ii) in more detail. It requires that $\int \vec{E}\cdot\vec{n}\,d\sigma$ taken across each electrode remains unchanged as the polarization is established. Now all of the charge distributions we will consider correspond to charge displacements, or polarizations, leaving the sample neutral. We will not be concerned with the injection of a charge, producing a deviation from neutrality. For analytical convenience, however, it will be convenient to generalize (ii), to include that case. If a net charge is introduced then the surface integral $\int \vec{E}\cdot\vec{n}\,d\sigma$ summed over the two electrodes must reflect the charge that has been introduced, and cannot vanish. Now we note that (ii) as defined above, corresponds to "floating" electrodes. The value of $\int \vec{E}\cdot\vec{n}\,d\sigma$ for each electrode is that determined by the dipole field in free space. There may be a local image charge pattern arising from a point dipole, since the electrode is an equipotential surface, but the net charge induced on each electrode by a dipole vanishes. The natural generalization of this: The lines of force emanating from a charge head equally for the two electrodes, as they would be in free space. Thus $\int \vec{E}\cdot\vec{n}\,d\sigma$ for the two electrodes changes equally as a charge is introduced into the sample. Figure 1 gives the potential distribution for a sheet dipole. Figure 1(a) corresponds to BC (i), whereas Fig. 1(b) corresponds to BC (ii). It is clear that the constant-potential boundary condition [Fig. 1(a)] corresponds to no change in the *space average* field, and therefore to no interaction with the positive host lattice charge (within the jellium model, where that constitutes a uniform positive charge). Let us, for the moment, assume that this is the BC implicity assumed by Sham, since he does not mention an interaction between the electron polarization and the positive host lattice.

In calculating the rate of momentum delivered by the field to the electron gas, Sham takes the total electron density in the alloy and multiplies it by a field value, without regard to the spatial variation of the electric field.[11] Now the actual electric field can be thought to differ from its average value in view of the presence of terms of the form shown in Fig. 1(a). Can electron-electron interactions, due to nonuniformities of this form, affect the net rate of momentum delivery to the volume involved? Normally, one thinks of electron-electron interactions as momentum conserving. That is obviously true, however, only in an infinite space, otherwise we must take boundary conditions into account. The dipole layer shown in Fig. 1(a) is, after all, accompanied by image charges on the electrodes.

Consider the interaction of two charged planes, one close to the left end of the specimen, the other in the middle of the sample. The field (not the potential) variations are shown in Figs. 2(a) and 2(b). The fields have been chosen so as to satisfy BC (i), i.e., $\int E\,dx = 0$. The charge densities in the

two planes are presumed to be equal, leading to the same discontinuity in E, in the two cases. If we consider the forces exerted by the two planes on each other, the field generated in Fig. 2(a), at a location in the middle of the sample, is relatively weak. The forces exerted by the charge planes on each other are opposite in direction, but far from equal in magnitude. Thus if we assume BC (i), the sum of the forces that the two charge planes exert on each other does not vanish.

Consider the contrasting situation associated with BC (ii). Here the fields emanating from a charge plane are independent of the plane's location, and only a function of the charge density in the plane. Thus the total force that two planes exert on each other vanishes, and electron-electron interactions can be ignored. We can, therefore, choose BC (ii) and cause electron-electron interactions to vanish, or else BC (i) and eliminate the interaction with the positive jellium host lattice; but we cannot make both effects disappear simultaneously. (In the case of a torus we presumably can make the two effects disappear simultaneously.)

While this author's earlier papers have not explicitly discussed the alternative possible boundary conditions, we have consistently invoked BC (ii). We have repeatedly used the equation $E = D - 4\pi P$, in analogy with dielectric theory. Here D is the field due to the charges on the electrodes E is the actual space-average field, and $4\pi P$ represents the change in field due to the dipole formation, assuming BC (ii). Sham's calculation goes on to include two further errors. (We again remind the reader that we are discussing the terminated sample, and that this may not be the best way to interpret Sham's equations.) If the electron-electron interactions produce no momentum change, then the field used to multiply the total electron density must be the field arising from charges outside the specimen (E_0 in Ref. 2, rather than E_T). Sham, however, uses the field used in Ohm's law, i.e., the space average field. Further, in calculating the direct force Sham again uses

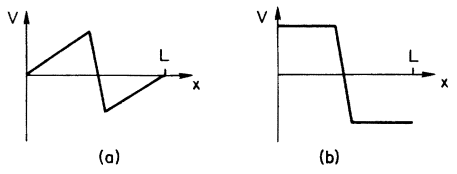

FIG. 1. Potential (V) due to a sheet dipole layer. (a) For constant potential difference, or short-circuited, electrodes. (b) For floating, or open-circuited electrodes. L is the distance between electrodes.

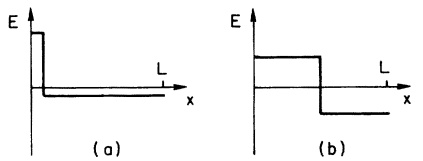

FIG. 2. Field distributions due to a charged sheet, between electrodes held at a constant potential difference. (a) for sheet near left end electrode. (b) for sheet halfway between electrodes.

the space average field, which includes the extra field due to the residual resistivity dipoles. But here, once again, only the field due to external charges (E_0) should be invoked, the effects of momentum transfer from the electron gas are, after all, already included in the momentum balance consideration which is the main point of Sham's Sec. III E.

The discussion, so far, has focussed on Ref. 3. While we have argued with the validity of Sham's derivation, it is clear that the agreement of his result, in his Eq. (3.30), with Refs. 1 and 2 is not just a typographical coincidence. Sham stresses that the distinction between ρ, the alloy resistivity, and ρ_0, the resistivity of the pure metal, arises from carrier density changes, as well as from changes in scattering action. By contrast Ref. 4 makes no allowances for the effect of carrier density changes on the resistivity. In fact, as pointed out very clearly by Turban *et al.*, they invoke Matthiessen's rule. Turban *et al.* also omit boundary condition questions.

Reference 4 consists of two, somewhat separate discussions. The initial portions of Ref. 4 enlarging upon a point made by Flynn[12] show that the effective charge, Z^*e, which determines the electromigration force equals the total charge which moves with the lattice defect, when that lattice defect is moved by nonelectrical forces, in the absence of an applied field.

Turban *et.al.* then go on, in their Sec. (2.4), to some essentially unrelated momentum balancing considerations. They engage in some initial considerations, in which they separate "bound" electrons from "free" electrons, with the implication that screening electrons are "bound" and that the number of electrons "free to reach the end of the crystal" must be equal to that in the pure host metal. This decomposition between free and bound electrons is baffling in the case of a repulsive impurity, or in the case where an attractive impurity is screened by an accumulation of mobile charges. The distinction causes difficulty, however, even in the case where the screening elec-

trons are actually bound to an attractive impurity. The original conduction electron density distribution is disturbed, even in that case. Are these spatial disturbances part of the "bound" charge distribution or part of the "free" charge? Near an attractive impurity potential, even after complete screening by bound states, the conduction band states are still perturbed in such a way that the electrons move faster, near the impurity. The higher velocities at the Fermi surface lead to increased current fluctuations. Through fluctuation-dissipation theory this in turn implies a higher conductivity. That is what we have called, "carrier density modulation," or "conductivity modulation" in earlier papers. Its effects are included in the Das-Peierls theorem derivation given in Refs. 1 and 2. It is an effect which is not compatible with Matthiessen's rule.[11] Most of the microscopic theories of electromigration, aside from those generated by the authors of Refs. 1 and 2, seem to have had difficulty with the carrier density modulation concept. While the quantative treatment of three-dimensional carrier density modulation has only been given in a very approximate and physical discussion,[11] the basic concept, as exemplified by the one-dimensional case,[13] is elementary.

Turban *et al.* then go on to provide a force balance argument for the "bound" charge. In this balance they omit: (i) The interaction of the polarized bound charge distribution with the positive host lattice; (ii) The interaction of this bound charge distribution with the lattice vibrations. The importance of this term depends on the exact definition of "bound" charge, i.e., whether the "bound" charges carry current or not.

In their subsequent Eq. (33), Turban *et al.* state that the total force on the impurity consists of that transmitted via the bound and free electrons supplemented by that exerted directly by the field. In this latter field related term they invoke the space average field. We have already criticized that point in connection with our discussion of Ref. 3. Turban *et al.* then go on to invoke a force balance for the "free" electrons. In this they once again ignore the interaction of local polarizations with the uniform positive host lattice (or else they ignore the electron-electron interactions, depending on their choice of boundary conditions). Subsequently, they invoke Matthiessen's rule. As already pointed out, that eliminates any remaining possible effects of carrier density changes on the resistivity.

The final section of Ref. 4, "Thermodynamic Analysis," once again disregards some of the points we have emphasized, but we will not take the space, here, to make that case in further detail.

We would like to include here some supplementary comments, which we hope will clarify some concepts related to the preceding discussions but which are not strictly essential to the discussion of Refs. 3 and 4.

(i) Carrier density modulation (or conductivity modulation) results from the acceleration of a nonuniform electron gas. The existing discussions of that[9] have emphasized the case where the spatially nonuniform momentum generation term is balanced by lattice vibration scattering. Actually, however, the treatment of Ref. 11 does not really invoke the presence of lattice vibration scattering.

(ii) Residual resistivity dipoles are inevitably accompanied by the interference terms first discussed by Bosvieux and Friedel.[14] This has been pointed out particularly clearly by Schaich,[15] and also by this author.[13] Conductivity modulation, as discussed in Ref. 11, should also be accompanied by interference terms which have not yet been discussed.

ACKNOWLEDGMENT

The author has been stimulated by a number of discussions with B. Bell and L. Sham, though Sham and I may not have arrived at complete agreement. To the extent that I have been critical of Sham's paper,[3] I also have to accept a share of the responsibility for that material. Reference 3 was generated in close interaction with me, and when first written, I agreed with the validity of Sec. III E of Ref. 3, as it stands.

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