Fundamental questions in the theory of electromigration

by A. H. Verbruggen

The theory of electromigration is focused on the force acting on a lattice defect in a metallic sample that carries an electric current. Much work has been done to obtain a better understanding of the underlying physical mechanisms. The force has been calculated numerically for defects in several metals, and a qualitative agreement with the experiments has often been found. There are, however, still discussions about the relevance of certain contributions to the force. These originate from conceptual difficulties related to 1) the nature of the screening of the electric field at the site of an impurity by the conduction electrons and 2) the existence and significance of inhomogeneities in the electric field and current flow near an impurity. This paper provides a review of the basic models and of questions which still exist in the theory of electromigration. The relevance of these questions is illustrated by results of experimental work on the electromigration of H in V, Nb, and Ta.

1. Introduction

When a strong electric current is passed through a metallic sample, the diffusional motion of impurities and/or vacancies is biased in a direction along or opposite to the current flow. This phenomenon is called electromigration [1], and recent technological interest arises from its manifestation as a cause of failure in integrated circuits. In the context of solid-state physics, theoretical studies of electromigration are focused on describing the various contributions to the driving force. This work revealed a number of still-existing fundamental conceptual problems which are important for a better understanding of electron transport in metals. For instance, a clear physical picture of the local electric field around an impurity in a current-carrying sample has not yet been given. This paper provides an overview of the basic models and questions in the theory of electromigration. In a simple picture, the driving force is split into two contributions: The first arises from the direct action of the external field on the charge of the migrating ion ("direct force"), the second from the scattering of the conduction electrons by the impurity or vacancy under consideration ("wind force"). Therefore,

$$\vec{F} = Z_b e\vec{E} + \vec{F}_{\text{wind}} = Z^* e\vec{E}, \tag{1}$$

where Z_b is the bare valence of the migrating defect with respect to the host lattice, e the elementary charge assumed to be positive throughout this article, and $\vec{E}$ the macroscopic (space-averaged) electric field. Z^* is called the effective valence of the defect. The wind-force contribution was ignored before the mid-fifties. Electromigration was called "electrolysis of metallic alloys." In 1954, in an unpublished internal IBM memorandum, Landauer derived an expression for the wind force by using momentum balance arguments. In the first part of this paper we discuss the development of the description of wind force.

The publication of a paper by Bosvieux and Friedel in 1962 [2] was the beginning of a strong disagreement about

the existence of the direct-force contribution. It was argued that the electric field at the site of, say, an interstitial impurity is screened by the conduction electrons. This controversy, which still exists, and further developments are described in the second part.

In order to provide a feeling about the experimental situation, as an intermezzo, results of electromigration measurements of hydrogen in V, Nb, and Ta are presented [3]. The results are regarded as a source of inspiration to look for field inhomogeneities. Landauer's residual resistivity dipole [4] and carrier density modulation [5] provide mechanisms for such inhomogeneities. These concepts are discussed in the fifth section. Finally, conclusions are drawn, and it is pointed out that electromigration theory is expected to benefit from insights obtained in the field of mesoscopic physics.

2. Wind force

The first published model, describing the interaction between conduction electrons carrying the electric current and the migrating ion, is the ballistic model of Fiks [6] and Huntington and Grone [7]. In this model it is assumed that the wind force on a lattice defect is given by the momentum transfer by the electrons per unit time as they are scattered by the defect. For an impurity in a free electron gas, this idea can be formulated as

$$\vec{F}_{\text{wind}} = \sum_{\vec{k},\vec{k'}} \hbar(\vec{k} - \vec{k'}) P_{\vec{k}\vec{k'}} f_{\vec{k}}(1 - f_{\vec{k'}}), \tag{2}$$

where $\vec{k}$ is the wavevector of the electrons and $P_{\vec{k}\vec{k'}}$ the transition probability given by the generalized Golden Rule; $f_{\vec{k}}$ is the shifted Fermi–Dirac distribution function

$$f_{\vec{k}} = f^0_{\vec{k}} + \left(-\frac{\partial f^0}{\partial \varepsilon}\right)(-e)\tau \vec{V}_{\vec{k}} \cdot \vec{E}, \tag{3}$$

where $f^0_{\vec{k}}$ is the Fermi–Dirac distribution in the absence of the electric field, e the elementary charge, τ the relaxation time of the electrons including all scattering mechanisms, $\vec{V}_{\vec{k}}$ the Fermi velocity, and $\vec{E}$ the macroscopic electric field. An evaluation of (2) using (3) leads to

$$\vec{F}_{\text{wind}} = \frac{n\tau}{n_i \tau_i}(-e)\vec{E}, \tag{4}$$

or

$$\vec{F}_{\text{wind}} = \frac{n\rho_i}{n_i \rho}(-e)\vec{E}, \tag{5}$$

where n (n_i) is the electron (impurity) density, τ_i is the transport lifetime corresponding to the scattering by the impurity under consideration, ρ is the total resistivity, and ρ_i is the contribution of the impurities to the resistivity. The wind force in this model is directed opposite to $\vec{E}$. $\vec{F}_{\text{wind}}$ is proportional to the residual resistivity ρ_i and inversely proportional to the total resistivity. The wind force is thus expected to decrease with increasing temperature. For a defect producing an increase in resistivity of 1 $\mu\Omega$-cm per at%, we find for, say, copper at room temperature

$$\frac{n\rho_i}{n_i \rho} \sim 50.$$

In free-electron-like metals the wind-force contribution is thus expected to be dominant. This is confirmed experimentally for some metals, but much smaller values for the wind force have also been found [1]. Cases where the wind force is in the same direction as the electric field are no exception either (see [1] and Section 4).

It is not clear how the ballistic model has to be extended if we want to go beyond the free-electron approximation. First, the momentum of a Bloch electron is not equal to $\hbar\vec{k}$, and second, it is not obvious how, in more complicated cases such as the scattering by clusters of atoms or by atom-vacancy complexes, the momentum transfer must be partitioned among defects and lattice. This problem is circumvented in the approach of Bosvieux and Friedel [2]. Due to the scattering of the electrons by a defect, the current flow near the defect is disturbed. This disturbance can be regarded as a current-induced local polarization of the charge distribution. In a stationary situation, the force exerted by the lattice defect on the electrons for maintaining these disturbances is equal to the force exerted on the defect by the polarized charge distribution. The wind force is then given by

$$\vec{F}_{\text{wind}} = \int n(\vec{r}) \left[-\frac{\partial V_b}{\partial \vec{R}_i}\right] d^3r, \tag{6}$$

where $n(\vec{r})$ is the actual electron density in presence of impurity and electric field, and V_b is the bare potential of the defect located at position $\vec{R}_i$. Within an independent-particle approximation, $n(\vec{r})$ around an impurity can be obtained by populating the electron-scattering states $\psi_{\vec{k}}(\vec{r})$ according to the shifted Fermi–Dirac distribution function $f_{\vec{k}}$ [Equation (3)]:

$$n(\vec{r}) = \sum_{\vec{k}} f_{\vec{k}} |\psi_{\vec{k}}(\vec{r})|^2. \tag{7}$$

For a justification of Equation (6), reference is usually made to the Born–Oppenheimer adiabatic approximation and/or to the Feynman–Hellman theorem. Equation (6) is also recovered in evaluations of Kubo's linear response formalism [8] for the driving force by Sham [9] and Schaich [10]. The importance of using the linear response formalism is that starting from a many-body statistical mechanical expression one is able to delineate in a systematic way the nature of the introduced approximations.

Equations (6) and (7) are the starting point of all calculations of the wind force in *real* metals. There are, however, rather subtle nuances, for instance in how self-consistency is achieved.

Sorbello [11] calculated the wind force for vacancies and impurities for virtually all free-electron-like metals within the pseudopotential formalism. The same technique was used by Genoni and Huntington [12] to study the anisotropy of Z^* in Zn. Lou et al. [13] used pseudopotential models of increasing sophistication to calculate the anisotropy of Z^* in Zn, Cd, and Mg. In general, the agreement between these theoretical results and experimental work is reasonable, especially in view of the sensitivity of the computations to the choice of form factor and the inaccuracy of some experimental data. Gupta [14, 15] evaluated Equation (6) within the muffin-tin formalism for calculating the wind force in noble and transition metals. The results for the noble metals seem satisfactory. In Nb, values for Z^* for vacancies and impurities are an order of magnitude too small. Brand and Lodder [16, 17] studied this system by using a finite cluster of muffin-tin potentials. In this model multiple scattering effects are preserved. Their results are satisfactory, although there is a marked dependence on cluster size. The same model was applied to migrating hydrogen interstitials in transition metals (see Section 4).

In spite of the reasonable agreement between theory and experiment, the situation is still not completely satisfactory, since questions remain about the validity of approximations made in the evaluations of Equation (6). Landauer [5] emphasized that certain contributions are ignored if we evaluate Equation (6) in the Born approximation, as discussed in Section 5. At present, full state-of-the-art calculations and more accurate experimental data are still highly desirable.

3. Direct force

Bosvieux and Friedel [2] presented the first nontrivial theory of the direct-force contribution in electromigration. Until then the direct force was assumed to be given by $Z_b e\vec{E}$ [see Equation (1)].

For a free-electron model *without* current and the electric field treated as a perturbation, Bosvieux and Friedel arrived (by using first-order stationary-state perturbation theory) at the following conclusions: 1) in the free-electron approximation, all ions (and all electrons) in a pure metal feel the electric field without screening; 2) an impurity in a metal does *not* feel *any* supplementary force with respect to the metal ions as a consequence of its different charge. This can be interpreted as meaning that any difference in valence of an impurity with respect to the metal atoms is screened by the conduction electrons and is ineffective for the direct force. As a consequence, there is no direct force on an interstitial impurity. These conclusions, drawn in 1962, were the beginning of a debate which is still proceeding. Gupta and coworkers [15] asserted in 1983 that the direct force is zero for a migrating ion in the saddle-point position. On the other hand, in 1986 Lou et al. [13] used the first term in Equation (1) for the direct force.

To illustrate the conceptual difficulty: For a neutral atom in an electric field, it is well known that the applied field at the nucleus is exactly compensated by a field due to the polarization of the electrons in bound states. It is also well known that an impurity in a metal is screened by the conduction electrons. However, it is not at all clear that the screening by conduction electrons is tied to the impurity in such a way that the electric field at the site of an interstitial impurity vanishes completely. Das and Peierls [18] argue that if this were true for, say, a proton in a metal, it would also hold for an electron, and a metal should not have any electric conductivity.

An argument used by workers who advocate the existence of the direct force is that eventual screening of the electric field by the electrons should follow from the expression for the electron–impurity interaction as used by Bosvieux and Friedel for the wind force, i.e., Equation (6). It is obvious that complete screening of the direct force cannot occur in the limit of weak scattering. The part of $n(\vec{r})$ that gives a nonzero contribution to the force is first-order in V or Z. Consequently, F_{wind} is of order Z^2, in contrast to the screening of Bosvieux and Friedel, which is first-order in Z.

Along the same line, the work of Das and Peierls [18], using the Boltzmann equation for describing the interaction between an ion and a free-electron gas carrying a current, or the work of Schaich [19], based on the density matrix formalism, should have yielded the screening of the direct force if it were present. This is not the case. Screening shows up in the final results only in the scattering potential of the impurity.

Recently Sorbello [20] succeeded in deriving a tractable expression for the direct force on a *strong* scatterer in a free-electron gas starting from the linear response mechanism. A model calculation for an s-scatterer shows the possibility of a significant reduction or enhancement of the direct force. Unfortunately, the theory is formulated in such a way that it is not clear (to the author) what the physical origin of this behavior is. Sorbello emphasizes that the "screening" in this theory is dynamical and is not present in a static polarization type of calculation like the one performed by Bosvieux and Friedel. This dynamical aspect is also present in Landauer's carrier density modulation (CDM) effect [5] (see Section 5).

In the context of this controversy, it is interesting to examine experimental work on electromigration; therefore, we consider the results of electromigration measurements of H in V, Nb, and Ta.

4. Electromigration of H in V, Nb, and Ta

The hydrides of the group-V transition metals are especially attractive for an experimental study of electromigration. Hydrogen occupies in these metals the tetrahedral interstitial sites, and the diffusion coefficient is very large ($D \sim 10^{-5}$ cm^2/s). This makes it possible to perform accurate

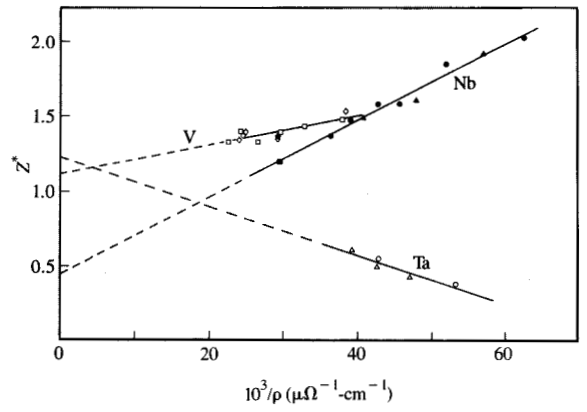

Figure 1

Effective valence Z^* against inverse resistivity for H in V, Nb, and Ta. The lines are least-squares fits of the expression $Z^* = Z_d + K/\rho$ (□, $VH_{0.0155}$; ◇, $VH_{0.0100}$; ●, $NbH_{0.0110}$; ▲, $NbH_{0.0115}$; ■, $NbH_{0.0103}$; ○, $TaH_{0.0085}$; △, $TaH_{0.0108}$).

Table 1 Parameters obtained from least-squares fits of the expression $Z^* = Z_d + K/\rho$ to electromigration data for H in V, Nb, and Ta.

	Z_d	K ($\mu\Omega$-cm)
V	1.11 ± 0.1	9.9 ± 1
Nb	0.44 ± 0.1	26 ± 2.5
Ta	1.23 ± 0.1	−16 ± 1.5

experiments over a wide temperature range well below the melting temperature of the metal.

From an electromigration experiment we obtain the effective valence Z^* defined in Equation (1). The temperature dependence of Z^* provides information about the various contributions to the driving force. In all Boltzmann-equation types of theories, the wind force is proportional to the relaxation time of the electrons, leading to $\vec{F}_{wind} = Ke\hat{E}/\rho(T)$ (K is a constant) [see Equation (5)]. We expect the temperature dependence of Z^* to be described by

$$Z^* = Z_d + K/\rho(T), \tag{8}$$

where Z_d is the temperature-independent direct-force contribution. Measured values for the effective valence Z^* for H in V, Nb, and Ta are plotted against inverse resistivity in **Figure 1**. The data are remarkably well described by Equation (8). The parameters obtained from least-squares fits of this relation are given in **Table 1**. For H in all three metals, Z_d is clearly different from zero. This cannot be understood without taking into account a temperature-independent direct-force contribution [21]. Particularly striking are the Ta results, which would require the wind force to increase with increasing temperature if no direct force were assumed. This would be at variance with all theoretical models of the wind force in electromigration published so far. It is especially interesting, however, that although Z_d for H in V is very close to the value +1 expected for a bare proton, in Nb and Ta sizable deviations from this value are found. This demonstrates in fact the complexity of the direct-force contribution.

As mentioned in the previous section, values for Z_d deviating from Z_b have been found by Sorbello in his theory on the direct force on a strong scatterer in a free-electron gas [20]. The results can also be interpreted as evidence for the existence of electric field inhomogeneities. In the analysis of the experiments, the field is assumed to be uniform and equal to its space-averaged value. Along this line we arrive at Landauer's CDM effect, which provides a mechanism for field inhomogeneities near lattice defects independent of the relaxation of the electrons. We discuss this in the next section.

The wind-force contribution in VH_x, NbH_x, and TaH_x shows a very interesting behavior. For H in V and Nb, the wind force is in the same direction as the direct force, whereas the opposite is true for H in Ta. Moreover, the magnitude of the wind-force contribution to the driving force is of the same order as the direct force. As mentioned in Section 2, for free-electron-like metals the wind force is often an order of magnitude larger. This shows that the role of the host lattice is very important. The small wind force is thought to be due to the compensation of contributions. Lodder and Brand [17] found in calculations of the wind force for H in V, Nb, Ta, Cu, and Pd a large compensation between contributions corresponding to different angular momentum numbers. Gupta et al. [15] ascribe a low value for the wind force for vacancies in Nb to cancellations between the contributions of various parts of the Fermi surface. In this spirit it is understandable that the sign for the wind force is not the same for all group-V transition metals, although they have a very similar electronic structure.

5. Field inhomogeneities

Landauer [5] has emphasized that when Equation (6) is evaluated within the Born approximation, two corrections should be taken into account. The first is the residual resistivity dipole (RRD) field, which makes a contribution to the wind force. The second, the carrier density modulation effect, provides a mechanism for a screening-like contribution to the direct force.

• *Residual resistivity dipole*

The basic idea behind the RRD is the following. Consider the introduction of an extra impurity in a current-carrying

metallic sample. Maintaining the current flow implies that the voltage across the sample, or the space-averaged electric field, goes up. It is very unlikely that the increase in field is uniform; it must be related to the position of the additional scatterer. Landauer studied this extra field in his 1957 paper [4]. By using a semiclassical analysis, he found that the charge around the scatterer is distributed in a dipolar way. The spatial average of the resulting dipole fields around the impurities is the same as that obtained by the usual approach, which does not explicitly consider the spatial variation.

The dipole field is non-oscillatory and is, since it is proportional to the cross section of the scatterer, of second or higher order in the potential.

Sorbello [22] has calculated the RRD field explicitly for a (strong) s-scatterer in jellium using partial-wave-scattering theory. In addition to other terms, a contribution is found which is exactly the dipolar distribution as obtained by Landauer if s-wave scattering is assumed in his expression.

In [23] it was stated that when the scattering state ψ is written as $\psi = \psi_0 + \psi_{sc}$, where ψ_0 is the unperturbed wavefunction, the RRD field corresponds to $|\psi_{sc}|^2$. However, as pointed out by Sorbello and Dasgupta [24] in their reference 42, one should also take into account terms corresponding to $\psi_0^*\psi_{sc}$. Gupta [25] noted recently that the RRD field for an s-scatterer as found by Sorbello [22] is entirely due to the $\psi_0^*\psi_{sc}$ contribution.

The significance of the RRD for the driving force in electromigration must be seen in relation to the work of Bosvieux and Friedel [2]. These authors used the Born approximation and calculated $n(\vec{r})$ to the first order in V. As mentioned above, the RRD field is of second or higher order in the potential and gives in the case of strong scattering a significant correction to the wind force. In later work (see, e.g., Schaich [10]) it was noted that there is no need for an order-by-order calculation of the corrections. The scattering problem can be solved exactly.

- *Carrier density modulation*

The origin of the carrier density modulation effect [5, 26, 27] is the increased (reduced) electron density in the immediate vicinity of a lattice defect with an attractive (repulsive) potential. Continuity of current then implies a change in velocity (or acceleration) of the carriers near the impurity. In a stationary situation this acceleration must be caused by a local electric field. Or, from another point of view, a change in conductivity near a defect implies a local change in electric field. For an attractive potential the CDM field is in the opposite direction from the background field. This means that the direct force on a migration impurity is reduced. Thus, the CDM effect provides an alternative screening mechanism for the direct force. The CDM effect is also important for resistance measurements.

Landauer [27] has derived a qualitative model for the effect of CDM on both the driving force in electromigration and the electrical conductivity. Sorbello and Dasgupta [24], using a density matrix technique, found in the lowest order in Z the same CDM effect on the conductivity. Sorbello's theory [20] on the direct force produces results similar to those of CDM, but at the moment it is not clear how the two are related.

The most serious appeal to the CDM effect has been made by Das and Peierls [28] and Landauer [29] in explaining what Landauer called the Das–Peierls electromigration theorem. By considering the momentum balance of the electrons in the presence of field impurities and background scattering, Das and Peierls found that the force on an ion (including the direct force) is given by

$$\vec{F} = -n_0(\Delta\rho/\rho)(-e)\vec{E}, \tag{9}$$

where n_0 is the electron density of the pure metal and $\Delta\rho$ is the change in resistivity caused by a defect density of one per unit volume. The resemblance of Equation (9) to Equation (5) for the wind force suggests that the direct force is removed by screening, contrary to the conclusions in Section 4. Das and Peierls and Landauer assert that this "screening" is in fact a manifestation of the CDM effect. These authors have speculated that the Das–Peierls electromigration theorem might have an extended range of validity because it is an exact relation between observable quantities. Plotting the results from Figure 1 as a function of $\Delta\rho(T)/\rho(T)$ shows that the force does not vanish as $\Delta\rho(T)/\rho(T) \to 0$, so the electromigration theorem does not give a correct description of the experimental data. This calls for a critical examination of the momentum balance of electrons in real metals.

6. Conclusions

Although quantitative calculations of the driving force in electromigration have been made which are in reasonable agreement with the experiments, it is at present not clear whether or not all relevant contributions have been taken into account correctly. Fundamental questions are the following:

1. To what extent is the applied field screened by the conduction electrons at the site of an impurity in a current-carrying sample?
2. What is the nature of the field inhomogeneities near a lattice defect; how are they described, and what is their relevance for electromigration?

Insight into these questions is of interest for the wide field of electron transport in solids.

Electromigration theory is expected to benefit from the field of mesoscopic physics [30]. Explanations of the Aharonov–Bohm effect and the universal conductance fluctuations observed in samples with dimensions in the submicron regime are based on the "Landauer formula" [4]. This expression relates the transmission of a potential barrier

to electrical resistance. In this approach the current is the causative source, and a field inhomogeneity around a scattering center arises as a natural ingredient.

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