

## Calculation of the electromigration wind force in Al alloys

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The electromigration wind force in various Al alloys is calculated using a Green's-function method for the calculation of the electronic structure. The influence of the environment of the jumping atoms is studied in detail in the Al-Cu alloy. Alloys of Al with  $3d$  and  $4sp$  alloying elements are studied systematically in order to investigate the relation between the electronic states of the alloying atom and the wind force. The study also includes several other alloys, which have been used in the past in attempts to increase electromigration lifetime. It is shown that the wind force on an Al host atom can be changed considerably by the presence of an alloying atom at particular positions near the jump path. This could be an additional contribution to the well-known decelerating effect of some alloying elements on electromigration in Al. [S0163-1829(99)01112-1]

### I. INTRODUCTION

Electromigration is the consequence of the interaction of an electric current and the atoms present in a metal sample. The result is a flux of atoms, which is proportional to the applied electric field or the current density. Observations of this effect for hydrogen in Pd date back to 1931,<sup>1</sup> and at first raised predominantly academic interest in the subject. However, with the use of thin Al lines as interconnects in electronic devices, electromigration became of interest also from a technological point of view.<sup>2</sup> The use of further miniaturized Al interconnects leads to larger current densities, which enhances electromigration and can therefore decrease the lifetime of a device.

Because of the proportionality of the driving force to the electric field  $\mathbf{E}$ , the driving force  $\mathbf{F}$  is usually characterized by the so-called effective valence  $Z^*$

$$\mathbf{F} = Z^* e \mathbf{E}, \quad (1)$$

where  $e$  is the elementary charge. There are two contributions to the driving force, which are called the direct force and the wind force. The direct force, which was the subject of intense debate,<sup>3</sup> is the result of a net charge of the atom. The estimates of this charge vary from the host valence charge, which is +3 in the case of Al, to zero. In most cases this direct force is outweighed by the wind force, which arises from momentum transfer from the current-carrying electrons to the atom. In the present paper a well-established quantum-mechanical expression<sup>4,5</sup> is used in order to calculate this force from first principles. The formalism has already been published elsewhere<sup>6</sup> and the main ingredients are addressed in Sec. II.

The atomic flux depends not only on the driving force, but also on the mobility of the atoms, as is clearly expressed in the Nernst-Einstein equation for the atomic drift velocity<sup>7</sup>

$$\mathbf{v} = \frac{D}{kT} \mathbf{F}, \quad (2)$$

with the diffusivity  $D = D_0 \exp(-Q/kT)$ , containing the frequency factor  $D_0$ , the activation energy  $Q$ , Boltzmann's constant  $k$ , and the temperature  $T$ .

An interesting observation, mainly because of its direct practical applicability, has been the fact that the addition of a few weight percent of Cu to an Al conductor reduces the electromigration-induced damage dramatically.<sup>8</sup> Even today the exact mechanism of this retardation is not fully understood. However, its occurrence led to attempts to alloy Al with many elements, some of which gave similar results.

One observation is that, when the Cu concentration exceeds a certain threshold, the flux of Al atoms is dramatically reduced.<sup>9</sup> Only after Cu is depleted, do the Al atoms start to drift. Considering Eq. (2) it seems that Cu must have an influence on either the activation energy  $Q$ , on the frequency factor  $D_0$ , or on the driving force through the effective valence  $Z^*$ . Although the most common explanations consider changes in the diffusion parameters  $Q$  and  $D_0$ , there is circumstantial evidence that the presence of Cu also affects the driving force.<sup>10</sup> Besides, it is well known that the atomic fluxes of host and alloying atoms are not independent.<sup>11</sup>

In the present paper the wind force is calculated on jumping alloying atoms and Al host atoms in the presence of alloying atoms. Results of these calculations are given in Sec. III, which are discussed in Sec. IV. Pure Al and the Al-Cu alloy are considered extensively in Secs. III A and IV A. Other alloying elements are considered in Secs. III B and IV B and in Secs. III C and IV C. Conclusions and further prospects are given in Sec. V.

### II. THEORY

The wind force on an atom at position  $\mathbf{R}$  is calculated using the well-established expression given by Bosvieux and Friedel<sup>4</sup> and Sorbello *et al.*<sup>5</sup> and

$$\mathbf{F}_{\text{wind}} = \sum_k f(k) \langle \psi_k | -\nabla_{\mathbf{R}V} | \psi_k \rangle, \quad (3)$$

in which the electric field induces a distribution  $f(k)$  of electrons, deviating from the Fermi-Dirac distribution  $f_{\text{FD}}$

$$f(k) = f_{\text{FD}}(E_k) - e\tau \mathbf{E} \cdot \mathbf{v}_k \delta[E(k) - E_F]. \quad (4)$$

Unlike the symmetrical part  $f_{\text{FD}}$ , the asymmetrical part, consisting of the current-carrying electrons at the Fermi surface, induces a force. The transport relaxation time  $\tau$  is inversely proportional to the resistivity.  $\mathbf{v}_k$  is the electronic velocity of an electron with band index and crystal momentum  $k \equiv (n\mathbf{k})$ ,  $v_1$  is the potential of the atom at position  $\mathbf{R}$  for which the wind force is calculated, and  $\psi_k$  is the electronic wave function. The Fermi-Dirac distribution and the relaxation times are the only quantities depending on temperature.

In order to calculate the wave function, we need to define the system in which the electrons move. We model the case of a dilute alloy, where only one defect region is embedded in a lattice of unperturbed host atoms. The defect region contains the jumping atom, a vacancy, and the atoms in its close environment. Using a Green's-function formalism the wave function for electrons in an alloy can be calculated, as described by Dekker *et al.*<sup>6</sup> In the following, the most important features of this calculation are described.

Both the alloy and the host wave functions are expressed in terms of local functions, centered at atomic position  $\mathbf{R}_n$ ,

$$\psi_k^{(\text{host})}(\mathbf{x} + \mathbf{R}_n) = \sum_L c_{knL}^{(\text{host})} R_L^{n,(\text{host})}(\mathbf{x}). \quad (5)$$

The wind force can be written in terms of the wave-function coefficients  $c_{k1L}$  for the jumping atom as

$$\mathbf{F}_{\text{wind}} = 2 \sum_k f(k) \text{Re} \sum_{Lm_1} c_{k1L}^* \mathbf{D}_{L,l+1,m_1} \sin(\eta_{l+1} - \eta_l) \times e^{i(\eta_{l+1} - \eta_l)} c_{k1l+1,m_1}, \quad (6)$$

where  $\eta$  are the phase shifts for scattering of electrons on potential  $v_1$ . A Lippmann-Schwinger equation relates the wave function of the host and the alloy and contains the host Green's function. In this basis the Green's function is represented by a matrix, which is calculated via a Brillouin-zone integration. This step is the most time consuming part of the calculations.

When the positions of the atoms in the alloy are chosen, the Green's-function matrix can be calculated. Then the positions in the defect region can be occupied by any desired atom or, formally, by any desired potential. Here the region consists of the nearest-neighbor shells of both the initial and the final positions of the jumping atom, as shown in Fig. 1. The defect region contains 20 lattice positions and the jumping atom is in the center of it. We stress again that in our calculations this defect is surrounded by unperturbed host atoms. Of course, in reality the environment is perturbed by the presence of alloying atoms and vacancies in the defect region. However, perturbations from these distant atoms are expected to be very small. In order to illustrate this, we refer to Fig. 2 in Sec. III A. There the wind force on atom 20 in Fig. 1 is shown in two situations, i.e., for an Al atom (solid arrow) and for a Cu atom (dashed arrow) at position 1. The change of the wind force due to this replacement turns out to

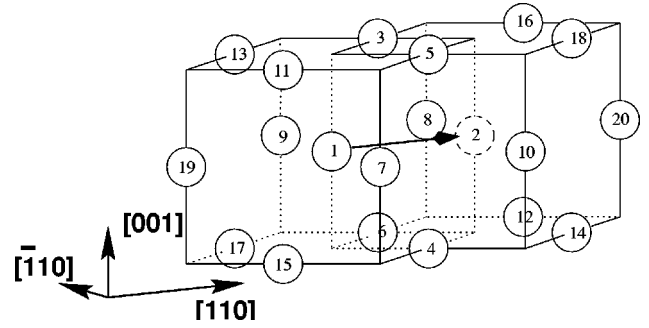


FIG. 1. The defect region. Atom 1 is the jumping atom, 2 denotes the original position of the vacancy. The remaining labeled atoms are individually included in the calculations.

be negligible and the effect of replacement of unperturbed Al atoms by Al atoms perturbed by charge transfer is also likely to be negligible.

Assuming that the jumping atom moves along a straight line from lattice site 1 to site 2, its position  $\mathbf{R}$  can be written as

$$\mathbf{R} = \mathbf{R}_1 + s(\mathbf{R}_2 - \mathbf{R}_1), \quad (7)$$

where  $s$  runs from 0 to 1. The wind force varies along the path and can be written as a function of this variable  $s$ . In our calculations the jumping atom is the only one which does not occupy a host-lattice position. Slight shifts of the positions of other atoms could also be taken into account, but this is not done here.

The wind force is not necessarily parallel to the applied current, which implies that the wind valence is a tensor  $\bar{Z}_{\text{wind}}(s, T)$ . However, the wind force can do work only in the direction of motion. The work done during the jump in direction  $\hat{s}$  can be written as

$$W = \frac{a}{\sqrt{2}} \int_0^1 ds [\hat{s} \cdot \rho(T) \bar{Z}_{\text{wind}}(s, T)] \cdot e \mathbf{j}, \quad (8)$$

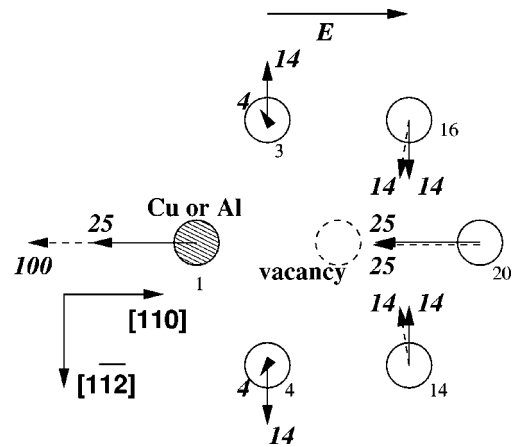


FIG. 2. The wind force on the nearest neighbors of the vacancy in the  $(1\bar{1}1)$  plane for an electric field in the  $[110]$  direction. The solid arrows refer to the situation with an Al atom at position 1, the dashed arrows to a Cu atom at that position. The small numbers label the atoms for reference in the text. The large numbers give the magnitude of the forces in eV/cm for a current density of  $1 \text{ MA/cm}^2$ .

where the electric field  $\mathbf{E}$  is expressed as the product of the electrical resistivity  $\rho(T)$  and the current density  $\mathbf{j}$  and in which  $a/\sqrt{2}$  is the length of the jump path, proportional to the lattice constant  $a = 4.05 \text{ \AA}$ . This work induces a bias in the diffusion process.

Because of the proportionality of the wind valence to the transport relaxation time  $\tau$ , which follows from Eqs. (1), (3), and (4), we can define the temperature-independent vector

$$\mathbf{K}(s) = \hat{s} \cdot \rho(T) \bar{Z}_{\text{wind}}(s, T), \quad (9)$$

which depends on  $s$ , parametrizing the position along the path. If all atoms, except the jumping atom, are Al atoms, this vector is parallel or antiparallel to the direction  $\hat{s}$  of the jump due to the symmetry of the system. When the symmetry is broken by the presence of an alloying atom, this vector also has a component orthogonal to  $\hat{s}$ .

As we will see in Sec. III A, this component influences the probabilities for particular jumps and therefore the detailed migration process. However, because of the symmetry of the fcc lattice such a jump is always compensated by a jump in the other direction and therefore does not lead to a net atom transport. The integral of the component of  $\mathbf{K}(s)$  in the path direction  $\hat{s}$  along the path

$$K = \int_0^1 ds \hat{s} \cdot \mathbf{K}(s) \quad (10)$$

gives a bias resulting in the actual migration and results in the wind part of the effective valence, which can be written as

$$Z^*(T) = Z_{\text{direct}} + \frac{K}{\rho(T)}. \quad (11)$$

Both  $Z_{\text{direct}}$  and  $K$  are independent of temperature. In most cases the wind force will be characterized by  $K$ , which is given in  $\mu\Omega \text{ cm}$ .

### III. RESULTS

#### A. Pure Al and the Al-Cu alloy

In this section several effects of the presence of Cu near a jumping Al atom will be presented. First, the wind forces on atoms next to a vacancy are shown when one of the Al atoms is replaced by a Cu atom. In this way it will be seen how the Cu atom influences its environment and how far its influence reaches. Second, the work done by the wind force, which is determined by the vector  $\mathbf{K}$ , will be considered.

The wind force on atoms next to a vacancy in the  $(\bar{1}\bar{1}1)$  plane is shown in Fig. 2 for the indicated direction of the electric field and two different situations. Only the components in the  $(\bar{1}\bar{1}1)$  plane are shown; the components of the force perpendicular to the plane are small in all cases. In the first situation, indicated by the solid arrows, no Cu is present near this vacancy. The electric field is parallel to the jump path of the atom labeled 1. The length of the arrows is proportional to the magnitude of the forces, which are also given explicitly in  $\text{eV/cm}$  for a current density of  $1 \text{ MA/cm}^2$ . As already suggested in Sec. II, the wind force is not necessarily parallel or antiparallel to the electric field. Most remarkable

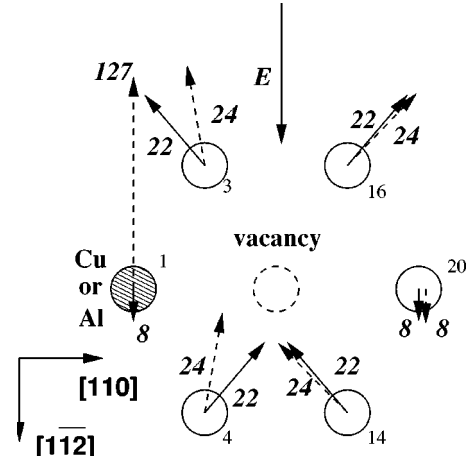


FIG. 3. Analogous to Fig. 2, but for an electric field in the  $[\bar{1}\bar{1}2]$  direction.

in this respect is the force on the atoms labeled 3, 16, 14, and 4. These forces are smaller than the forces on atoms 1 and 20, but they are almost perpendicular to the electric field. This does not agree with the notion of a scalar wind valence, which is based on the assumption that the force on the atom is a direct result of the scattering of the applied current. Such an assumption would lead to a force opposite to the electric field.

In the second situation, atom 1 is replaced by a Cu atom. This is indicated by the dashed arrows. The force on the Cu atom has risen to four times the force on an Al atom in the same position (dashed arrow not drawn to scale). The replacement significantly reduces the magnitude of the forces on atoms 3 and 4 and changes their direction. The influence on the other atoms is much smaller because of the larger distance to the Cu atom. Nevertheless the atoms 16 and 14 notice the presence of the Cu atom through a slight change in the direction of the force.

In Fig. 3 both situations are shown again, but the electric field is now rotated by  $90^\circ$ . Again the direction of the force on the atoms 3, 16, 14, and 4 is not parallel to the electric field. In this case the Cu atom does not have a large influence on the magnitudes of the forces on the Al atoms, but it changes the direction on atoms 3 and 4 considerably. The forces on the atoms 16, 20, and 14 are changed only slightly. A Cu atom at position 1 feels again a force much larger (by a factor of 15) but opposite to an Al atom. This can be explained by dividing the force into two parts: one as a result of the applied current and one as a result of the local current arising from scattering of the applied current to the environment of the jumping atom.<sup>12</sup> The original applied current is not scattered by a host Al atom at a lattice position, and therefore the force due to the scattered current dominates. On the other hand, a Cu atom scatters both currents and the calculations show that the applied current dominates.

In pure Al the vector  $\mathbf{K}$  is antiparallel to the jump direction  $\hat{s}$  along the entire path. This means that work is done only by the component of the applied current in the direction  $\hat{s}$ . For a current density of  $1 \text{ MA/cm}^2$  in that direction the work done along the path is  $W = -1.10 \text{ } \mu\text{eV}$ . This corresponds to  $K = -39 \text{ } \mu\Omega \text{ cm}$  or a wind valence of about  $-15$  at room temperature ( $\rho = 2.65 \text{ } \mu\Omega \text{ cm}$ ). In the paper

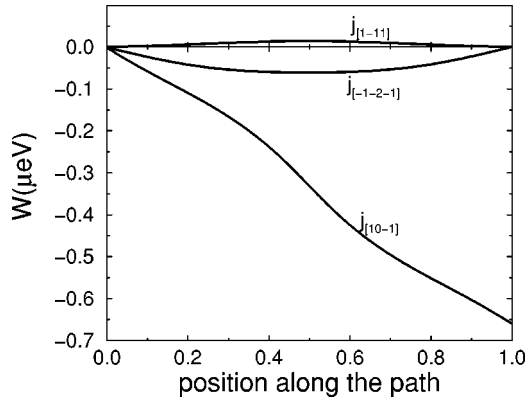


FIG. 4. The work done during a jump of atom 2 in Fig. 2 by an applied current density of  $1 \text{ MA/cm}^2$  in three orthogonal directions  $[10\bar{1}]$ ,  $[1\bar{2}1]$ , and  $[1\bar{1}1]$ .

by Dekker *et al.*<sup>6</sup> erroneously a value of  $K = -29 \mu\Omega \text{ cm}$  was reported. For a Cu jump,  $\mathbf{K}$  is also antiparallel to the jump path and a value of  $K = -97 \mu\Omega \text{ cm}$  is found, which is a factor of 2.5 larger than for Al.

For the jump of the Al atom labeled 3, with a Cu atom at position 1,  $\mathbf{K}$  is not parallel to the  $[10\bar{1}]$  jump direction. The work done along the path for three orthogonal directions of the current is displayed in Fig. 4. A current of  $1 \text{ MA/cm}^2$  in the  $[10\bar{1}]$  direction of the path decreases the energy by  $0.658 \mu\text{eV}$ . A current in the direction orthogonal to  $[10\bar{1}]$  has a somewhat more subtle effect. The total work done along the path is zero, and therefore no net transport is expected. On the other hand, the wind does net work on the first half of the path and thereby slightly changes the barrier for this jump. However, this is expected to be of minor importance.

The work done during all jumps in Fig. 2 for three orthogonal directions of the current is shown in Table I. During a jump of the Cu atom at position 1 and a jump of Al atom 1, work is only done by the wind force arising from the  $[110]$  component of the current. The work done during a jump of atom 20 is about 10 percent larger than when the Cu atom is absent. With and without the Cu atom, jumps of atoms 3 and 4 are made more difficult by a current in the  $[110]$  direction. The effect, however, is reduced by the presence of Cu. In contrast, a jump of the atoms 16 and 14 is made easier by the presence of Cu. The work done during

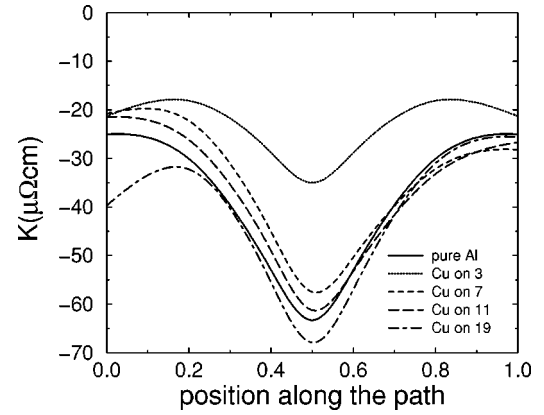


FIG. 5. Variation of  $K$  for a host Al atom with a Cu atom on one of the positions in the perturbed cluster. The labels of the positions correspond to those in Fig. 1.

jumps of the atoms 3, 16, 14, and 4 is decreased by the presence of Cu for a current in the  $[1\bar{1}2]$  direction.

A current in the  $[1\bar{1}1]$  direction does not do work on any atom in pure Al jumping in the  $(1\bar{1}1)$  plane. The presence of Cu changes this situation. The jump of atom 16 is inhibited by the Cu atom, while a jump of atom 14 is supported. However, the work done by the current in the  $[1\bar{1}1]$  direction is one order of magnitude smaller than the work done by currents in the  $(1\bar{1}1)$  plane.

Finally, the variation of the component parallel to the direction of the jump of the vector  $\mathbf{K}$  is shown in Fig. 5 for a Cu atom at positions 3, 7, 11, and 19 in Fig. 1. This is the component leading to the bias in the motion of the atoms. As already seen above, the presence of a Cu atom at position 3 reduces the wind force in the saddle point by a factor of 2. The average over the path decreases by 40%. Together with the atoms at the positions 4, 5, and 6 it forms a ‘‘gate’’ through which the jumping atom passes.

### B. 3d and 4sp impurities

The wind valences of the 3d and 4sp alloying atoms in Al averaged along the jump path are shown in Fig. 6 together with experimental values for their impurity resistivities.<sup>13</sup> Both quantities are a result of scattering of the current carrying electrons and have large values for the impurities with a half-filled  $d$  shell. The maximum wind valence of  $-470$  at

TABLE I. Work (in  $\mu\text{eV}$ ) done by the wind force due to a current density of  $1 \text{ MA/cm}^2$  in three orthogonal directions during the first and second halves of the jumps in Fig. 2.

Atom	$\hat{j} = \frac{1}{\sqrt{2}}[110]$		$\hat{j} = \frac{1}{\sqrt{6}}[1\bar{1}2]$		$\hat{j} = \frac{1}{\sqrt{3}}[1\bar{1}1]$	
	$W$	$W_{\text{pure Al}}$	$W$	$W_{\text{pure Al}}$	$W$	$W_{\text{pure Al}}$
1	-2.782	-1.100	0	0	0	0
3	-0.329	-0.550	-0.571	-0.953	0	0
16	0.623	0.550	-0.887	-0.953	-0.020	0
20	1.194	1.100	0	0	0	0
14	0.623	0.550	0.887	0.953	0.020	0
4	-0.329	-0.550	0.571	0.953	0	0

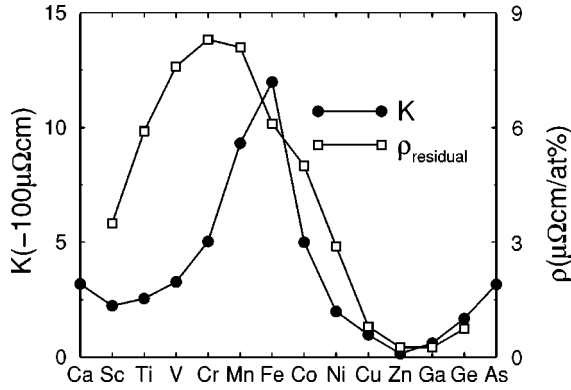


FIG. 6. Calculated wind valences  $K$  for jumping  $3d$  and  $4sp$  alloying atoms and experimental values for their residual resistivity (Ref. 13).

room temperature is found for Fe. The impurity resistivity follows a similar peak, but it is less pronounced and has shifted from Fe to Cr.

The contributions  $K_{sp}$ ,  $K_{pd}$ , and  $K_{df}$  according to Eq. (6) in the initial position for the  $3d$  transition metals are shown in Fig. 7. The  $pd$  and  $df$  contributions lead to the peak for the elements with a half-filled  $d$  band. This suggests an important role for the  $d$  states.

Figure 8 shows  $K$  for the jumping Al atom, when position 3 is occupied by a  $3d$  or  $4sp$  alloying atom. The effective valence is reduced significantly by alloying elements with a half-filled  $d$  band, which feel the largest wind force themselves. For example, the presence of Fe at such a position reduces the wind force on Al by more than 80%. As can be seen from the open circles in Fig. 8, the presence of an alloying atom at the considered position does not always lead to a suppression of the wind force. The alloying elements Sc, Ti, V, and Cr lead to an increase of the wind valence at the initial position by up to 30%. However, averaged over the migration path, the wind force is reduced.

### C. Other alloys

In this section, the wind force in Al alloys, which were used in earlier attempts to decrease the electromigration damage, are considered. In Sec. IV C, these results will be compared with experimental data, and their link with technologically interesting lifetime measurements will be investigated.

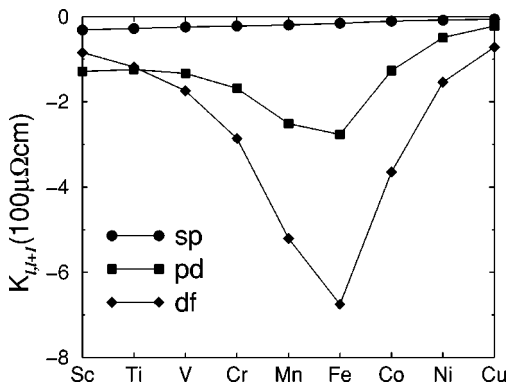


FIG. 7. Calculated wind valences  $K_{sp}$ ,  $K_{pd}$ , and  $K_{df}$  for the  $3d$  transition-metal alloying atoms in Al in the initial position.

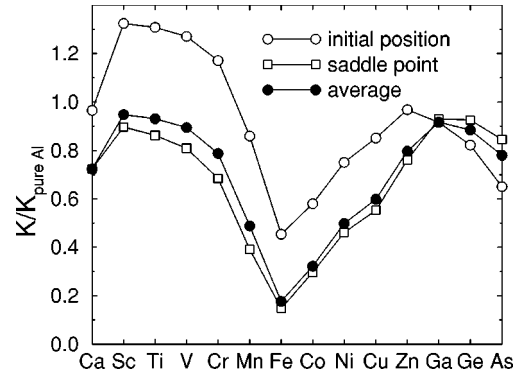


FIG. 8.  $K$  for Al in the initial and saddle-point position, and the average over the migration path with  $3d$  and  $4sp$  alloying atoms at position 3 in Fig. 1.

The results for  $K$  of the alloying atoms are shown in Table II. The calculated wind valence of Ag is about as large as for Cu, while the one of Au is much larger. An Au atom on position 3 also induces the largest reduction of the wind force on Al. The group-IVA metal alloying atoms have larger wind valences than the noble-metal atoms. On the other hand, their effect on the wind valence of a nearby jumping Al atom is small. Mg has a very small wind valence according to these calculations and also a small influence on the Al wind valence. Pd behaves similarly to the  $3d$  alloying atom Ni, which was treated in Sec. III B, just as Ag behaves like Cu. For Si a large wind valence and a small influence on a jumping Al atom are found.

## IV. DISCUSSION

### A. Cu

These calculations have shown that the addition of an alloying atom complicates the electromigration process considerably. It is seen that the wind force is not necessarily parallel to the electric field and current, which would be the

TABLE II. Wind valences for different alloying atoms in Al and wind valences of Al, when position 3 is occupied by an alloying atom  $K_{Al}^{alloy}$  normalized to the wind valence in pure Al  $K_{Al}^{pure}$ .

Alloying atom	$K(\mu\Omega\text{ cm})$	$K_{Al}^{alloy}/K_{Al}^{pure}$
Noble Metals		
Cu	-96	0.60
Ag	-102	0.58
Au	-373	0.34
Group IVA		
Ti	-255	0.93
Zr	-556	0.80
Hf	-398	0.83
Other alloying atoms		
Mg	-10	0.91
Pd	-167	0.51
Si	-186	0.90

case for a scalar wind valence. The wind force on an Al atom is changed due to the presence of a Cu atom near the migration path as shown in Figs. 2 and 3.

The work done by the wind force along the path supports particular jumps and can be used to characterize the wind force by the temperature-independent quantity  $\mathbf{K}$ , defined in Eq. (9). This vector is parallel to the jump path when the atoms in the environment of the jumping atom are host atoms. For the jump of an Al atom near a Cu atom these vectors also have other components. They lead to very subtle changes of the jump probabilities as shown in Fig. 4 and Table I. Therefore, the kinetics of the electromigration process is slightly changed. On the other hand, these probability changes for certain types of jumps do not induce migration because of symmetry.

The component leading to electromigration, given by Eq. (10), is used in the definition of the effective valence [Eq. (11)]. Our calculations give a value of  $K = -39 \mu\Omega \text{ cm}$  for pure Al and a 2.5 times larger wind valence,  $K = -97 \mu\Omega \text{ cm}$ , for Cu in Al. A Cu atom occupying position 3 in Fig. 1 reduces the wind valence of the Al atom jumping from position 1 by 40%.

It is interesting to consider the influence of these results on the measured fluxes of Al as well as Cu atoms. If Al jumps only with a Cu atom at position 3, the wind valence on Al is reduced by 40%. On the other hand, if the probability of finding a Cu atom in each of the 18 positions of Fig. 1 is 1%, the wind valence is only reduced by 2%. The influence of these changes in the effective valence therefore cannot be predicted in a straightforward way. One of the important parameters in the electromigration process is the binding energy between vacancies and Cu atoms. If this energy is large, Cu atoms and vacancies form pairs, which are difficult to separate. Then two types of jumps are frequent, jumps of Cu atoms and jumps of Al atoms with a Cu atom at position 3. This means that the suppression of the wind valence would be large. It is clear that the implications of these calculations can only be estimated in combination with all other parameters. This could possibly be done with a kinetic Monte Carlo study of the atomic transport process, which takes into account the effective valences calculated here. Work along these lines is currently in progress.

### B. 3d and 4sp alloying elements

The 3d transition-metal alloying atoms, as investigated in this section, all experience a larger wind force than Cu. The peak as a function of atomic number, which has been found experimentally<sup>13</sup> and theoretically<sup>14–17</sup> for the residual resistivity, is qualitatively also reproduced by the wind valence, but it is sharper and its position is shifted from Cr to Fe. These differences could be induced by the proximity of the vacancy or by the fact that the atom does not occupy a lattice position during the jump. Another possible origin of these differences could be the calculation procedure, which is discussed by comparing different calculations for the impurity resistivity.

Calculations in which the Fermi surface is approximated by a sphere tend to underestimate the resistivity.<sup>14–16</sup> However, a Green's-function method similar to the one used in the present paper, taking this anisotropy into account, still

leads to an underestimate.<sup>17</sup> For the alloying atoms with a large number of *d* electrons and 4*sp* alloying atoms, the agreement with experiment can be improved using a model procedure to account for the amount of charge transfer due to the alloying atom.<sup>18</sup> This procedure was found to be less suitable for alloying atoms with a small number of 3*d* electrons.<sup>17</sup> The charge transfer is not accounted for self-consistently in the present implementation of our method. It would be very interesting to calculate the electronic structure of these alloys self-consistently and see how this changes the wind force.

A central result of our calculations is that the presence of one of these alloying atoms near the path of a jumping Al atom can strongly reduce the wind force on Al. The largest reduction, e.g., 80% in the case of Fe, was found for impurities with a large wind valence. This conforms with the intuitive picture of a local current in the opposite direction of the applied current, induced by the alloying atom. It stands to reason that this induced current is large for strong scatterers.

### C. Other alloying elements

The results for the noble metals indicate that Au should have the largest influence on the electromigration properties of Al. The correlation between the wind valence of an alloying atom and the influence on the wind valence of a host atom found in Sec. III B is confirmed by the calculations for noble-metal alloying elements. However, experiments show that the addition of Ag or Au hardly changes the electromigration behavior.<sup>19</sup> This shows that the correlation between the driving force considered here and the macroscopic behavior is not so simple.

Alloying group-IVA elements are interesting for different reasons. Ti is often present in the substrate or in the passivation layer,<sup>20,21</sup> whereas the addition of a small amount of Zr to a Al-Cu alloy leads to a decrease of the lifetime.<sup>21</sup> As for the noble-metal alloying elements, their influence on the electromigration behavior cannot be explained using these calculations only.

The element Mg also reduces the electromigration kinetics.<sup>22,9</sup> Interestingly, the wind valence of Mg is very small, i.e.,  $K_{\text{Mg}} = -10 \mu\Omega \text{ cm}$ , which is even a factor of 4 smaller than the wind valence of a host Al atom. In the case of a very small or vanishing force, the alloying element is only transported due to the vacancy flux or, equivalently, the transport of host atoms. The wind valence obtained is interesting in view of recent Blech experiments showing that Mg migrates very slowly and seems to be left behind after the Al has moved.<sup>22,9</sup>

The alloying element Pd shows interesting features in combination with other alloying elements. The addition of a small amount of Pd in combination with V has been shown to improve the electromigration properties considerably.<sup>23</sup> Other experiments suggested that Pd inhibits electromigration in combination with hydrogen or Nb.<sup>21</sup> The wind force on Pd is larger than that on Cu. A Pd atom can decrease the wind valence of Al by a factor of 2.

It is interesting to finally consider Si in Al, because it is always present in electronic circuits. Its wind valence is about twice as large as for Cu, whereas it has hardly any effect on the wind valence of a nearby Al atom.

Overall, we must conclude that at present a simple correlation between our microscopic calculations and the macroscopic electromigration behavior cannot be established. Our calculations as they stand give only the driving force for particular jumps in the alloy, regardless of how often such a jump takes place. It is to be hoped that this information can in the future be linked with other modeling approaches, leading to a more thorough and quantitative understanding of alloying effects in electromigration.

## V. CONCLUSIONS AND OUTLOOK

Several effects of the addition of alloying elements to aluminum have been investigated theoretically by an *ab initio* method. Even in pure Al the wind valence has been found to be anisotropic. The average over all the resulting jumps leads to a measurable wind force, which is parallel (or antiparallel) to the applied current. This measurable component of the force is calculated for pure Al to be  $K = -39 \mu\Omega \text{ cm}$ , which agrees well with recent experiments.<sup>24</sup>

Our calculations show that most of the alloying elements have a larger wind valence than the host Al atoms. This is expected as the applied current is scattered by alloying elements, but not by host atoms. For Cu we find a wind valence which is 2.5 times larger than for Al. Almost all 3*d* alloying elements have a large wind valence, with Fe having the largest value, which is more than 30 times larger than for Al.

The wind valence for these atoms follows roughly the impurity resistivity, as expected. The wind valences for the 4*sp* alloying elements are rather low. Interestingly the wind valence for Zn, which has two 4*s* electrons, is found to be smaller than for Al. Such a small value is also found for Mg, which has two 3*s* electrons. The latter result is consistent with recent Blech-type experiments.<sup>22,9</sup>

The wind valence of the noble-metal alloying elements alone does not explain why Cu slows down the electromigration process whereas Ag and Au do not. The wind valence of Ag is about as large as the one of Cu, while the wind valence of Au is a factor of 4 larger.

A second alloying effect could be the change of the wind force on a host atom due to the proximity of an alloying atom. It turns out that this can increase as well as decrease the wind force and that these effects are more pronounced when stronger scatterers are added. This is consistent with the picture that local currents are induced, which alter the wind valence with respect to the situation without alloying atom.

The problem of how to link these results with electromigration measurements has been discussed. Our microscopic calculations, which give only the driving forces for particular jumps, would need to be combined with kinetic simulations in order to achieve a more quantitative understanding of alloying effects in electromigration.

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