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Generalized Friedel sum from Lloyd's formula

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Expressions for the generalized Friedel sum rule are derived which are applicable as a condition for charge neutrality at defects in metallic systems. These defects include the thoroughly studied substitutional impurity as well as the poorly explored interstitial impurity and an atom at the saddle-point position between two vacancies, being typical for substitutional electromigration. The expressions are derived starting from Lloyd's formula for the density of states of an arbitrary collection of scattering potentials.

I. INTRODUCTION

Lloyd's formula[1] for the density of states of an arbitrary collection of nonoverlapping scattering potentials in free space was derived in a period during which was investigated how far, as concerns the electronic properties, an infinite solid could be modeled by a finite collection of atoms.[2,3] An important application of this formula is found in later studies of dilute alloys.[4,5] Lehmann[4] derived a condition for charge neutrality in substitutional alloys starting from Lloyd's formula. This condition can be considered a straight generalization of the Friedel sum rule[6] for a single scatterer in free space. In fact, this simple rule follows immediately from Lloyd's formula if it is applied to a single scatterer.

While the electronic properties of dilute substitutional alloys have been studied extensively,[5,7] calculations for interstitial impurities are still in a rudimentary stage.[8,9] As far as the authors know, no calculation of a self-consistent interstitial-impurity potential has yet been carried successfully at the level of an early single-site calculation[10] for a substitutional impurity. This may be due to the fact that the interstitial Green's function suffers severely from free-electron poles, so that the necessary integrals over the Brillouin zone cannot be evaluated with sufficient accuracy.[11] Nevertheless, by now both Dingle temperatures[12] and electromigration properties[13] have been calculated for interstitial alloys. These calculations were done using constructed muffin-tin potentials, possibly adapted in a simple way in order to fulfill a generalized Friedel sum rule.

Interestingly, in these calculations an expression for this rule was used which was not derived explicitly but was written down as an analogy of Lehmann's expression[4] for a substitutional impurity. Since its derivation turned out to be by no means trivial, a first aim of the present paper is to remedy this situation. A second aim is inspired by developments in electronic structure calculations on special defects expected in the near future.[11,14] These defects are typical of electromigration in substitutional alloys.[14,15] An impurity migrating from its initial position to a neighboring vacancy is a very special defect indeed, particularly when it is halfway along its path. Judging it shallowly, it looks like a kind of interstitial-impurity defect, but regarding the usual description of a local defect in an otherwise perfect crystal it is quite different. In describing an impurity at a lattice site or at an interstitial position, all atoms in the defect have a one-to-one correspondence with an atom in the unperturbed host. This simple correspondence is broken for an atom halfway along its jump path, at the so-called saddle-point position. This atom has to be described with respect to *two* atoms of the host.

The second aim of this paper is to derive an expression for the generalized Friedel sum rule applicable to such a defect.

In Sec. II an expression is derived that is applicable to an interstitial-impurity defect, which includes possibly perturbed host atoms around the impurity by charge transfer and lattice distortion. In Sec. III a more general expression is derived which is applicable to the migrating-atom defect as well. Some comments and an outlook are given in Sec. IV.

II. THE INTERSTITIAL-IMPURITY CASE

The Lloyd formula for the electronic density of states at the energy E of a collection of nonoverlapping scattering potentials in free space is given by[16]

$$n(E)=n_0(E)+\frac{2}{\pi}\frac{d}{dE}\mathrm{Im}\,\mathrm{Tr}\ln T(E)$$
$$=n_0(E)-\frac{2}{\pi}\mathrm{Im}\,\mathrm{Tr}\left\{T\frac{dT^{-1}(E)}{dE}\right\}, \qquad (1)$$

in which $n_0(E)$ is the density of states of free electrons, and the system t matrix T can be expressed in terms of the t matrices t of the individual potentials, located at positions $\mathbf{R}_j$, and a purely geometrical structural matrix B as

$$(T^{-1})^{jj'}_{LL'}=t^{j\,-1}_{LL'}\delta_{jj'}-B^{jj'}_{LL'}\ . \qquad (2)$$

For spherical muffin-tin potentials the single-site t matrices are diagonal in the angular momentum label $L\equiv(lm)$, so that

$$t^{j}_{LL'}=t^{j}_{l}\delta_{LL'}\ , \qquad (3)$$

with

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$$t_l^j = -\frac{1}{\kappa}\sin\delta_l^j \exp(i\delta_l^j) , \tag{4}$$

in which $\kappa = E^{1/2}$ and δ_l^j are phase shifts.

The structural matrix B is given by

$$\begin{aligned} B_{LL'}^{jj'} &\equiv B_{LL'}(\mathbf{R}_{jj'}) \\ &= -4\pi\kappa i^{l-l'+1} \sum_{L''} i^{l''} C_{LL'L''} h_{L''}^{+}(\mathbf{R}_{jj'}) \end{aligned} \tag{5}$$

in which $C_{LL'L''}$ are Gaunt coefficients, $\mathbf{R}_{jj'} \equiv \mathbf{R}_j - \mathbf{R}_{j'}$, $h_L^+(\mathbf{r}) \equiv h_l^+(\kappa r) Y_L(\hat{\mathbf{r}})$, and $h_l^+(x) \equiv j_l(x) + i n_l(x)$, the latter spherical Hankel function being the standard combination of spherical Bessel and Neumann functions in scattering theory. The form (1) is slightly different from Lloyd's original form,[1] but it is the correct form according to Lehmann.[4] This has been confirmed[16] by explicit integration of the basic operator form

$$n(E) = -\frac{2}{\pi} \mathrm{Im}\, \mathrm{Tr}\, G(E) , \tag{6}$$

$G(E)$ being the Green's-function operator $(E^+ - H)^{-1}$ of the system.

The Lloyd formula (1) has two major advantages compared with the operator form. First, it is a matrix form, the labels jL of the matrices being composed of a site label j and the angular momentum label L. Second, the energy derivative occurs explicitly in it, so that the integrated density of states $N(E)$ can be read from (1) and (2) straightforwardly:

$$N(E) = N_0(E) - \frac{2}{\pi} \mathrm{Im}\, \mathrm{Tr} \ln(t^{-1} - B) . \tag{7}$$

Although this expression was derived originally for a finite collection of scattering potentials in free space, it can be applied to an infinite collection as well. If free space is interpreted as the muffin-tin zero in a muffin-tin representation of the crystal, the crystal density-of-states expression is shown[17] to reduce explicitly to the Lloyd formula. Therefore we will apply expression (7) to the interstitial-impurity case, though in the slightly rewritten form

$$N(E) = N_0(E) + \frac{2}{\pi} \mathrm{Im}\, \mathrm{Tr} \ln t - \frac{2}{\pi} \mathrm{Im}\, \mathrm{Tr} \ln(1 - tB) . \tag{8}$$

The interstitial-impurity case is characterized by one additional scattering potential in an interstice of the host, being surrounded by perturbed host atoms. This perturbation will include charge-transfer differences and position changes $\Delta_j = \mathbf{R}_j - \mathbf{R}_j^h$ due to lattice distortion. Due to the one-to-one correspondence of atoms, the host atom position label j also can be used in the dilute alloy for the perturbed host atoms, but the presence of the interstitial implies one more position, to be labeled by p. So in subtracting from expression (8) a similar expression for the host,

$$\begin{aligned} N(E) - N^h(E) = {} & \frac{2}{\pi} \mathrm{Im}\, \mathrm{Tr} \ln t - \frac{2}{\pi} \mathrm{Im}\, \mathrm{Tr} \ln t^h \\ & - \frac{2}{\pi} \mathrm{Im}\, \mathrm{Tr} \ln(1 - tB) \\ & + \frac{2}{\pi} \mathrm{Im}\, \mathrm{Tr} \ln(1 - t^h B^h) , \end{aligned} \tag{9}$$

one is faced with two problems. One manifest problem in reducing the right-hand side of (9) is that the third and fourth terms cannot be combined due to the different size of the two matrices. This comes from the extra position label p in the third term. Another problem is related to an additional difference between B and B^h due to lattice distortion. The first problem can be solved by enlarging the host matrix in the fourth term of (9) to the size of the alloy matrix in as natural a manner as possible. This is done by putting an empty sphere in the interstice labeled by p, having a t matrix equal to zero of course. The correspondingly enlarged matrix is related to the original matrix by

$$(1 - t^h B^h)^{pj,pj'} = \begin{bmatrix} 1 & 0 \\ -t_j^h B^{h,jp} & (1 - t^h B^h)^{jj'} \end{bmatrix} . \tag{10}$$

This equality is in matrix notation for the angular momentum labels. In view of the equality

$$\mathrm{Tr} \ln A = \ln \det A \tag{11}$$

holding for an arbitrary square matrix A the contribution of the matrix at the left-hand side of (10) to the fourth term in (9) is equal to the contribution of the original matrix, which stands in the right-bottom corner of the right-hand side of (10). Note that this trick would have failed if the form (7) would have been used in deriving the difference (9), instead of the form (8). The inverse t matrix $t^{h^{-1}}$ cannot be extended this way.

The lattice distortion problem can be remedied relatively simply, because matrices B and B^h, using a well-known expansion property,[1] are related by

$$\begin{aligned} B^{nn'} = B(\mathbf{R}_{nn'}) &= B(\mathbf{R}_{nn'}^h + \Delta_n - \Delta_{n'}) \\ &= J(\Delta_n) B^{h,nn'} J(-\Delta_{n'}) . \end{aligned} \tag{12}$$

The position label $n = pj$ as in (10), combining the interstitial and host-atom positions. The matrix $J(\Delta)$ takes care of the lattice distortion,[18] and is defined like the matrix $B(\mathbf{R}_{jj'})$ in Eq. (5), but the spherical Hankel function in (5) has to be replaced by a spherical Bessel function divided by $-i\kappa$. The matrix J has the property

$$J(\mathbf{x}) J(-\mathbf{x}) = 1 . \tag{13}$$

Applying (12) and (13) to the third term of (9), one writes

$$\begin{aligned} \mathrm{Tr} \ln(1 - tB) &= \mathrm{Tr} \ln[J(\Delta) J(-\Delta) - t J(\Delta) B^h J(-\Delta)] \\ &= \mathrm{Tr} \ln(1 - \tilde{t} B^h) , \end{aligned} \tag{14}$$

in which[19]

$$\tilde{t} = J(-\Delta) t J(\Delta) , \tag{15}$$

and use has been made of the invariance of the trace operation for the cyclic order in a matrix product.

One more step is required in order to be able to reduce Eq. (9) to a manageable form. By adding and subtracting a term $(2/\pi)\mathrm{Im\,Tr}\ln B^h$ in the right-hand side of (9), this equation, for E equal to the Fermi energy E_F, obtains the form

$$Z^{\mathrm{Friedel}}=\frac{2}{\pi}\mathrm{Im\,Tr}\ln t-\frac{2}{\pi}\mathrm{Im\,Tr}\ln t^h +\frac{2}{\pi}\mathrm{Im\,Tr}\ln \mathcal{G}-\frac{2}{\pi}\mathrm{Im\,Tr}\ln \mathcal{G}^h\ , \tag{16}$$

in which the left-hand side is equal to the valence difference $N(E_F)-N^h(E_F)$ between alloy and host system and

$$\mathcal{G}^h=B^h\frac{1}{1-t^hB^h}\ . \tag{17}$$

The alloy matrix $\mathcal{G}$ can be expressed in terms of $\mathcal{G}^h$ as

$$\mathcal{G}=B^h\frac{1}{1-\tilde{t}B^h}=B^h\frac{1}{1-t^hB^h-(\tilde{t}-t^h)B^h}=\mathcal{G}^h+\mathcal{G}^h\Delta t\,\mathcal{G}\ , \tag{18}$$

with

$$\Delta t=\tilde{t}-t^h\ . \tag{19}$$

Iteration of (18) leads to

$$\mathcal{G}=\mathcal{G}^h+\mathcal{G}^h\Delta t\frac{1}{1-\underset{\cdot\cdot}{\mathcal{G}}^h\Delta t}\mathcal{G}^h \tag{20}$$

and

$$\mathcal{G}\mathcal{G}^{h^{-1}}=1+\mathcal{G}^h\Delta t\frac{1}{1-\underset{\cdot\cdot}{\mathcal{G}}^h\Delta t}\ . \tag{21}$$

In these latter equations, dots are used to indicate that the range of the labels is restricted to the position labels of the defect, for which $\Delta t\neq 0$. Using (4), (11), and (21) in rewriting (16), the following form arises for the generalized Friedel sum:

$$Z^{\mathrm{Friedel}}=\frac{2}{\pi}\sum_{n,l}(2l+1)(\delta_l^n-\delta_l^{h,n})-\frac{2}{\pi}\arg\det(1-\underset{\cdot\cdot}{\mathcal{G}}^h\Delta t)\ . \tag{22}$$

This completes the derivation of the expression for an interstitial impurity, used in the past[12,13,20] without having a proper deviation. Lehmann's expression (3.13) for a substitutional impurity has exactly the form of (22), and the latter form reduces to Lehmann's expression by restricting the range of the label $n=pj$ to the host positions j. However, it should be realized that Lehmann's expression did not account for lattice distortion. The insight about how to include this effect came later.[18,20]

We end this section by showing that $\underset{\cdot\cdot}{\mathcal{G}}^h$ is equal to a familiar Brillouin-zone integral:

$$\begin{aligned}\mathcal{G}^h_{nn'}&=\left\langle n\left|B^h\frac{1}{1-t^hB^h}\right|n'\right\rangle\\&=\langle n|B^h+B^ht^hB^h+\cdots|n'\rangle\\&=B^h_{nn'}+\sum_{jj'}B^h_{nj}\left\langle j\left|\frac{1}{t^{h^{-1}}-B^h}\right|j'\right\rangle B^h_{j'n'}\\&\equiv B^h_{nn'}+\sum_{jj'}B^h_{nj}\mathcal{T}^h_{jj'}B^h_{j'n'}\ .\end{aligned} \tag{23}$$

By the last member the host t matrix $\mathcal{T}^h$ is defined. In the internal summation in (23) only lattice labels j remain, because $t_p^h=0$. The corresponding Fourier-transformed expression is

$$\mathcal{G}^h_{nn'}=B^h_{nn'}+\frac{1}{\Omega_{\mathrm{BZ}}}\int^{\mathrm{BZ}}d^3k\,e^{i\mathbf{k}\cdot\mathbf{R}_{nn'}}b^n(\mathbf{k})\times M^{-1}(\mathbf{k})\tilde{b}^{n'}(-\mathbf{k})\ . \tag{24}$$

Here the integral goes over a Brillouin zone, with volume Ω_{BZ}, the wiggle denotes the transpose matrix, and the b matrix is defined by

$$b^n(\mathbf{k})=\sum_j\exp(-i\mathbf{k}\cdot\mathbf{R}_{nj})B^h_{nj}\ , \tag{25}$$

with the sum running over all lattice sites, and $M(\mathbf{k})$ is the Korringa-Kohn-Rostoker (KKR) matrix

$$M(\mathbf{k})=t^{h^{-1}}-b(\mathbf{k}) \tag{26}$$

containing $b(\mathbf{k})=b^j(\mathbf{k})=\tilde{b}^j(-\mathbf{k})$ for j being an arbitrary lattice site. Expression (24) can be reduced to simpler forms[21] if either n or n', or both indices, refer to lattice positions, but always a Brillouin-zone integration remains to be performed. It is interesting to note that in Eq. (14) of Oppeneer and Lodder,[21] pertaining to two *different* interstitial positions, the first term in (24) is missing. In practice this has never led to errors, since only one interstice was present, and the B matrix is zero for equal position labels by definition.

III. AN EXPRESSION FOR A GENERAL DEFECT

Even for a general defect in a dilute alloy, it will be possible to indicate a region inside which the number, positions, and character of the scattering potentials are different compared with the host situation, and outside which one finds essentially an unperturbed host situation. This makes it attractive to work with cluster t matrices, one for the defect region,

$$T_{00}=J^{0\cdot}(t^{-1}-B_c)^{-1}J^{\cdot 0}\equiv J^{0\cdot}T^{\cdot\cdot}J^{\cdot 0}\ , \tag{27}$$

and one for the corresponding region in the host,

$$T^h_{00}=J^{0\cdot}(t^{h^{-1}}-B^h_c)^{-1}J^{\cdot 0}\equiv J^{0\cdot}T^{h,\cdot\cdot}J^{\cdot 0}\ . \tag{28}$$

The size of the matrices $t^{-1}-B_c$ and $t^{h^{-1}}-B^h_c$ can be different depending on the number of scatterers in the defect region. The position vector labeled by 0 is taken equal for the defect and host cluster, and can be considered to be chosen somewhere inside the cluster region. The matrix $J^{0\cdot}=J(\mathbf{R}_0-\mathbf{R}_{\cdot})$ has already been defined in

Sec. II, but the range of the right position label denoted by the dot can be different for the two clusters, because the dot positions run over the scatterers in the cluster. The subscript at the B matrices serves to indicate that their site labels run over the dot positions only. Expressions (27) and (28) are standard multiple-scattering results.

In view of the above, the alloy and host can be described by matrices of the same size, the position labels running over $\mathbf{R}_0$ and all unperturbed host positions $\mathbf{R}_i$. The matrices to be substituted in the integrated density of states expression (7) are equal as far as the B matrix is concerned, and differ as for t^{-1} only for the cluster position label 0. So a first step in deriving a condition for charge neutrality is to write

$$\begin{aligned} Z^{\text{Friedel}} &= N(E_F) - N^h(E_F) \\ &= -\frac{2}{\pi}\operatorname{Im}\operatorname{Tr}\ln \mathcal{T}_c^{-1} + \frac{2}{\pi}\operatorname{Im}\operatorname{Tr}\ln \mathcal{T}_c^{h^{-1}} \\ &= \frac{2}{\pi}\operatorname{Im}\operatorname{Tr}\ln \mathcal{T}_c \mathcal{T}_c^{h^{-1}} \ . \end{aligned} \tag{29}$$

The host t matrix

$$\mathcal{T}_c^h \equiv (t_c^{h^{-1}} - B)^{-1} \tag{30}$$

carries a subscript c to indicate the cluster character of one of the t matrices in it, and to distinguish it from the regular host t matrix $\mathcal{T}^h$, expressed in terms of the single-site t matrices only and occurring in Eq. (23). The corresponding alloy t matrix can be developed as

$$\begin{aligned} \mathcal{T}_c = \frac{1}{t_c^{-1} - B} &= \frac{1}{t_c^{h^{-1}} - B + t_c^{-1} - t_c^{h^{-1}}} \\ &= \mathcal{T}_c^h - \mathcal{T}_c^h \Delta(t_c^{-1}) \mathcal{T}_c \ . \end{aligned} \tag{31}$$

Similarly to what was done for the $\mathcal{G}$ matrix in Sec. II, iteration of (31) gives

$$\mathcal{T}_c = \mathcal{T}_c^h - \mathcal{T}_c^h \Delta(t_c^{-1}) \frac{1}{1 + \mathcal{T}_c^h \Delta(t_c^{-1})} \mathcal{T}_c^h \tag{32}$$

and

$$\mathcal{T}_c \mathcal{T}_c^{h^{-1}} = 1 - \mathcal{T}_c^h \Delta(t_c^{-1}) \frac{1}{1 + \mathcal{T}_c^h \Delta(t_c^{-1})} \ . \tag{33}$$

The structure of this matrix product to be substituted in (29) is similar to the product $\mathcal{G}\mathcal{G}^{h^{-1}}$ of Sec. II in the sense that only the cluster position label contributes. The reason is that $\Delta(t_c^{-1}) = 0$ for all labels except the label 0. So (29) can be shown to reduce to

$$Z^{\text{Friedel}} = -\frac{2}{\pi}\operatorname{Im}\operatorname{Tr}\ln[1 + \mathcal{T}_{c,00}^h (T_{00}^{-1} - T_{00}^{h^{-1}})] \ . \tag{34}$$

This is already a largely simplified form, because the size of the matrices is determined by the range of the angular momentum label of the cluster t matrices. Still it has two disadvantages. The first is that no small cutoff value for this cluster's angular momentum exists, in contrast to what holds for regular muffin-tin potentials. However, this might not be considered too serious a disadvantage in the present days[22] of multiple-scattering theory beyond the muffin-tin approximation. A more serious disadvantage is is that the way in which (34) has been derived is of limited validity for practical reasons. This is due to the implicit assumption made above, that the defect region can be treated as a separate scatterer. Only for very small muffin-tin radii can a defect enveloping sphere be chosen that is not overlapping[23] with the other atomic scatterers, if it can be chosen at all. The very defect we have in mind in this section is an oblong one corresponding to an electromigration jump for an atom to a neighboring vacancy, in which only ellipsoidal symmetry is left. The same problem was faced earlier.[15] Fortunately, it can be solved by making explicit the position labels of the scatterers inside the cluster, without being faced with so-called near-field corrections.[22] We will show that this is possible for expression (34) as well. In fact, regarding recent results[24,25] in formulating multiple-scattering theory in terms of t matrices of space-filling cell potentials, this is rather interesting. We think that earlier proof by one of the present authors[15] and proof given below can be interpreted as explicit demonstrations of the validity of the space-filling cell-potentials description, in which no near-field corrections occur. As a by-product the first proof will show that expression (34) as it stands is not yet general enough.

A. An elaboration of expression (34)

Use will be made of the intimate relation between the two host t matrices $\mathcal{T}^h$ and $\mathcal{T}_c^h$, defined by Eqs. (23) and (30), respectively. These matrices describe the same system, but in $\mathcal{T}^h$ all atoms are accounted for individually, while in $\mathcal{T}_c^h$ some of them, the ones in the defect cluster region, are treated collectively by the cluster t matrix T_{00}^h defined in (28).

Stated this way, one will be ready to believe that, for the diagonal site-0 matrix element occurring in (34), the following equality holds:

$$\mathcal{T}_{c,00}^h = J^{0\cdot} \mathcal{T}_{\cdot\cdot}^h J^{\cdot 0} \ . \tag{35}$$

Again the range of the dot position labels is restricted to the defect region. This equality holds, but its proof is rather tricky and will be postponed to Appendix A. In view of the forms (27) and (35), one has to develop the product

$$J_h^{\cdot 0} \frac{1}{J_a^{0\cdot} (t^{-1} - B_c)^{-1} J_a^{\cdot 0}} \equiv F^{\cdot 0} \ , \tag{36}$$

and a similar product found by substituting (28), for a further elaboration of (34). The h and a labels are added in order to remind one of the possibly different range of the dot position labels in the host and alloy defect regions, respectively. Some care is required in manipulating (36), because the J matrices are not square matrices and do not have an inverse in the usual sense. For further help, we define two other J matrices by

$$J_{ha}^{\cdot\cdot} \equiv J_h^{\cdot 0} J_a^{0\cdot} \tag{37}$$

and

$$J_{aa}^{\cdot\cdot} \equiv J_a^{\cdot 0} J_a^{0\cdot} \ . \tag{38}$$

In order to prevent confusion we remind the reader of the definition of the J matrices. If the B matrix in (5) is denoted by $B(h^{+})$ in abbreviated notation, a J matrix is given by $J=iB(j)/\kappa$. Using the properties of such matrices, it becomes known that (37) and (38) are not merely definitions, but the left-hand sides are the outcome of the elaboration of the complete internal L summations in the right-hand sides. For the sake of clarity we show this property for Eq. (37) by writing all labels explicitly:

$$J_{ha,LL'}^{j_c^h j_c}=\sum_{L''} J_{LL'}^{j_c^h 0} J_{L''L'}^{0 j_c} \ , \tag{39}$$

in which j_c^h and j_c indicate the positions of the scatterers in the host and alloy defect regions. Now we give the main steps in the manipulation of (36). First the inverse matrix at the left-hand side is brought to the right-hand side:

$$J_h^{\cdot 0}=F^{\cdot 0}J_a^{0\cdot}(t^{-1}-B_c)^{-1}J_a^{\cdot 0} \ . \tag{40}$$

Subsequently this equation is multiplied from the right by $J_a^{0\cdot}$, thereby generating the matrices $J_{ha}^{\cdot\cdot}$ and $J_{aa}^{\cdot\cdot}$. The latter square matrix is positive definite and can be inverted. After this one reads from (40) the equality

$$J_{ha}^{\cdot\cdot}J_{aa}^{\cdot\cdot\,-1}(t^{-1}-B_c)=F^{\cdot 0}J_a^{0\cdot} \ . \tag{41}$$

The matrix $F^{\cdot 0}$ is made explicit as follows:

$$F^{\cdot 0}=J_{ha}^{\cdot\cdot}J_{aa}^{\cdot\cdot\,-1}(t^{-1}-B_c)J_{aa}^{\cdot\cdot\,-1}J_a^{\cdot 0} \ , \tag{42}$$

which is seen immediately if the latter equation is multiplied by $J_a^{0\cdot}$ from the right. If the same manipulations are carried out for the term in (34) containing the host cluster t matrix, one finds

$$F_h^{\cdot 0}=(t^{h^{-1}}-B_c^h)J_{hh}^{\cdot\cdot\,-1}J_h^{\cdot 0} \ . \tag{43}$$

This can be read simply from (42), in which all a labels have to be replaced by h labels. After this, for (34) one finds

$$Z^{\text{Friedel}}=-\frac{2}{\pi}\operatorname{Im}\operatorname{Tr}\ln\{1+J_h^{0\cdot}\mathcal{T}^h_{\cdot\cdot}[J_{ha}^{\cdot\cdot}J_{aa}^{\cdot\cdot\,-1}(t^{-1}-B_c)J_{aa}^{\cdot\cdot\,-1}J_a^{\cdot 0}-(t^{h^{-1}}-B_c^h)J_{hh}^{\cdot\cdot\,-1}J_h^{\cdot 0}]\} \ . \tag{44}$$

As a final step the matrix in (44) is multiplied by $J_{hh}^{\cdot\cdot\,-1}J_h^{\cdot 0}$ from the left, and by $J_h^{0\cdot}$ from the right. It will be shown in Appendix B that the outcome of (44) is insensitive to such an operation. In this way we finally find

$$Z^{\text{Friedel}}=-\frac{2}{\pi}\operatorname{Im}\operatorname{Tr}\ln\{1+\mathcal{T}^h_{\cdot\cdot}[J_{ha}^{\cdot\cdot}J_{aa}^{\cdot\cdot\,-1}(t^{-1}-B_c)J_{aa}^{\cdot\cdot\,-1}J_{ah}^{\cdot\cdot}-t^{h^{-1}}+B_c^h]\} \ . \tag{45}$$

This expression can be evaluated straightforwardly by present computational means. The host t matrix reduces to a Brillouin-zone integration like (24):

$$\mathcal{T}_{jj'}^h=\frac{1}{\Omega_{\text{BZ}}}\int^{\text{BZ}} d^3k\, e^{i\mathbf{k}\cdot\mathbf{R}_{jj'}}M^{-1}(\mathbf{k}) \ , \tag{46}$$

and the J matrices are calculated routinely. However, it will soon become clear that expression (45) is not yet general enough, covering only a limited set of cases.

First, equations (45) and (22) should reduce to the same form for the case of a substitutional impurity. This can be shown simply indeed. The relation between host matrices $\mathcal{G}^h$ and $\mathcal{T}^h$ is

$$\mathcal{G}^h=-t^{h^{-1}}+t^{h^{-1}}\mathcal{T}^h t^{h^{-1}} \ . \tag{47}$$

Forgetting for the moment about lattice distortion, the matrix in (22) can now be written as

$$\begin{aligned}1-\mathcal{G}^h(t-t^h)&=1+(t^{h^{-1}}-t^{h^{-1}}\mathcal{T}^h t^{h^{-1}})(t-t^h)\\ &=t^{h^{-1}}[1+\mathcal{T}^h(t^{-1}-t^{h^{-1}})]t \ .\end{aligned} \tag{48}$$

The matrix in (45) reduces to

$$1+\mathcal{T}^h(t^{-1}-t^{h^{-1}}) \tag{49}$$

because without lattice distortion $J_{ha}\rightarrow J_{hh}$, $J_{aa}\rightarrow J_{hh}$ and $B_c\rightarrow B_c^h$. Regarding property (4), the factors $t^{h^{-1}}$ and t in (48) cancel exactly the phase-shift term in (22), by which (22) and (45) are shown to be equivalent for a substitutional impurity. It is just a matter of some additional algebra to show the equivalence in the presence of lattice distortion as well.

Now we show that expression (45) does not cover the case of an interstitial impurity. Neglecting lattice distortion, matrices $J_{ha}^{\cdot\cdot}$ and $J_{ah}^{\cdot\cdot}$ then differ from matrix $J_{aa}^{\cdot\cdot}$ only in that one row or column, respectively, is missing, namely the one corresponding to the position of the interstitial. This means that, through the product $J_{ha}^{\cdot\cdot}J_{aa}^{\cdot\cdot\,-1}$, being almost the complete unity matrix and therefore consisting only of Kronecker δ's, the matrix $t^{-1}-B_c$ contributes only its lattice-position columns. Its column for the interstitial is projected out.

In view of this one might wonder how general expressions (34) and (45) are. They cover the substitutional case, but defects containing more atoms than the corresponding host region are apparently excluded. However, the projection property does not hold for defects containing fewer atoms. So expression (45) is expected to be applicable in the electromigration case, particularly if the migrating atom at or around the saddle-point position is described by one t matrix. This can be done either by neglecting contributions of the remaining parts of the vacancies at its initial and final positions, or by including their contributions in the full local t matrix, which of course is nondiagonal in the angular momentum. This would mean that, in spite of its deficiencies, expression (45) could serve the second aim mentioned in Sec. I. Still it is worthwhile to discuss Eq. (34) further, because in its derivation no approximations were made. This will be done in Sec. II B.

B. A general defect

Equation (34) obtains a form which is applicable to a general defect after substitution of the equality

$$\mathcal{T}^h_{c,00}=T^h_{00}+T^h_{00}\mathcal{G}^{hc}_{00}T^h_{00}\ , \tag{50}$$

in which

$$\mathcal{G}^{hc}_{00}=\sum_{ii'}\mathbf{B}^{0i}\mathcal{T}^h_{ii'}\mathbf{B}^{i'0}\ . \tag{51}$$

This equality follows simply from (30) in the same way (47) could be derived. It can be seen that, after rewriting (47) slightly, it attains to the same form as (50). The internal matrix $\mathcal{T}^h$ in (51) should carry the subscript c, but for the position labels i outside the defect region it is shown in Appendix A that $\mathcal{T}^h_c=\mathcal{T}^h$. Substitution of (50) into (34) leads to

$$Z^{\text{Friedel}}=\frac{2}{\pi}\text{Im}\,\text{Tr}[\ln T_{00}-\ln T^h_{00}-\ln\{1-\mathcal{G}^{hc}_{00}(T_{00}-T^h_{00})\}]\ , \tag{52}$$

a form which is remarkably similar to form (22), regarding equality (11) and the form of the terms with phase shifts as it occurs in (16).

Now we are going to rewrite (52) in terms of matrices carrying position labels of individual sites, similarly to the step from Eqs. (44) to (45). For the third term matrices $J_s^{\cdot 0}$ and $J_s^{0\cdot}$ are used, the dot labels of which may refer to other positions than do the dot labels of alloy and host clusters. The reason for this will readily become clear. One obtains

$$Z^{\text{Friedel}}=\frac{2}{\pi}\text{Im}\,\text{Tr}[\ln T^{\cdot\cdot}-\ln T^{h,\cdot\cdot}-\ln\{1-(\mathcal{G}^{hc,\cdot\cdot}_{sa}T^{\cdot\cdot}J^{\cdot\cdot}_{as}-\mathcal{G}^{hc,\cdot\cdot}_{sh}T^{h,\cdot\cdot}J^{\cdot\cdot}_{hs})J^{\cdot\cdot\,-1}_{ss}\}]\ , \tag{53}$$

in which, e.g.,

$$\mathcal{G}^{hc,\cdot\cdot}_{sa}=J_s^{\cdot 0}\mathcal{G}^{hc}_{00}J_a^{0\cdot}=\sum_{ii'}\mathbf{B}_s^{\cdot i}\mathcal{T}^h_{ii'}\mathbf{B}_a^{i'\cdot} \tag{54}$$

is precisely a host Green's function matrix of the form required in the description of electromigration[15] [see Eq. (18) in that paper]. Equation (53) could be applied as it stands, but it appears profitable in practice to write the contribution of the single-site t matrices to its first two terms explicitly as follows:

$$\text{Im}\,\text{Tr}(\ln T^{\cdot\cdot}-\ln T^{h,\cdot\cdot})=\text{Im}\,\text{Tr}\{\ln t-\ln t^h-\ln(1-\mathbf{B}_c t)+\ln(1-\mathbf{B}^h_c t^h)\}\ . \tag{55}$$

Now it becomes interesting to see in which way (53) with (55) covers special cases. First we consider the interstitial case, again forgetting about lattice distortion for convenience. The s-dot positions are to be chosen equal to the alloy-cluster dot positions, and the host-cluster matrix $T^{h,\cdot\cdot}$ in the third term of (53) is to be enlarged to the alloy-cluster size, in the same way as was done in Eq. (10). Combining (55) with (53), one obtains

$$Z^{\text{Friedel}}=\frac{2}{\pi}\text{Im}\,\text{Tr}\left[\ln t-\ln t^h-\ln(1-\mathbf{B}_c t)+\ln(1-\mathbf{B}^h_c t^h)-\ln\left\{1-\mathcal{G}^{hc,\cdot\cdot}_{aa}\left[T^{\cdot\cdot}-t^h\frac{1}{1-\mathbf{B}_c t^h}\right]\right\}\right]\ . \tag{56}$$

Enlarging similarly the matrix $1-\mathbf{B}^h_c t^h$ in the fourth term of (56), and combining this term, now containing the matrix $1-\mathbf{B}_c t^h$, and the third term with the last term, it is found that

$$Z^{\text{Friedel}}=\frac{2}{\pi}\text{Im}\,\text{Tr}\left[\ln t-\ln t^h-\ln\left\{\frac{1}{1-\mathbf{B}_c t^h}\left[1-\mathbf{B}_c t-\mathcal{G}^{hc,\cdot\cdot}_{aa}\left[t-t^h\frac{1}{1-\mathbf{B}_c t^h}(1-\mathbf{B}_c t)\right]\right]\right\}\right]\ . \tag{57}$$

Three more equalities have to be used in order to observe the desired result. The first two are

$$\frac{1}{1-\mathbf{B}_c t^h}(1-\mathbf{B}_c t)=1-\frac{1}{1-\mathbf{B}_c t^h}\mathbf{B}_c\Delta t\ , \tag{58}$$

in which $\Delta t=t-t^h$, and

$$1+t^h\frac{1}{1-\mathbf{B}_c t^h}\mathbf{B}_c=\frac{1}{1-t^h\mathbf{B}_c}\ , \tag{59}$$

the latter being most easily derived by a series expansion of the inverse matrices. The third equality is

$$\frac{1}{1-\mathbf{B}_c t^h}\mathcal{G}^{hc,\cdot\cdot}_{aa}\frac{1}{1-t^h\mathbf{B}_c}=\mathcal{G}^{h,\cdot\cdot}_{aa}-\mathbf{B}^c\frac{1}{1-t^h\mathbf{B}_c}\ , \tag{60}$$

which was derived earlier[15] in a slightly different form [see Eq. (A9) in that paper]. Using (58)–(60) in rewriting (57), one finds

$$Z^{\text{Friedel}}=\frac{2}{\pi}\text{Im}\,\text{Tr}[\ln t-\ln t^h-\ln(1-\mathcal{G}^{h,\cdot\cdot}_{aa}\Delta t)]\ , \tag{61}$$

which is completely equivalent to the earlier form (22), obtained in a much different way. Although a migrating atom at the saddle-point position cannot be described as an interstitial, the techniques employed above to enlarge matrices by adding sites with t matrices equal to zero can be applied in the electromigration-defect case as well. One just has to enlarge the host-cluster matrices in (53) with (55) in the same way, just adding a $t^h=0$ site at the saddle-point position; but in addition one has to enlarge the defect-cluster matrices by adding two $t=0$ sites, one at the initial position of the migrating atom and one at its final position. The s label in (53) now refers to one more position compared with the h label, and two more positions compared with the a label. But in this way the expression for the generalized Friedel sum describing the migrating-impurity case obtains the same form as (61), the $\mathcal{G}$ matrix now carrying a subscript ss instead of aa. It will be a matter of actual numerical calculations to find out which of the form works better, (61) or the original form (53).

At present the task of this paper is complete. It will have become clear that Eq. (53) is the powerful form covering all conceivable cases. More specialized forms like (61) can be derived from it. The reason why form (34), being an exact form, becomes truly general only after the substitution of identity (50), requires further attention, and will be the subject of the next paper.[26] It will turn out that the correct general form always contains a Green's-function matrix, which fact is reducible to the presence of the system's Green's-function operator $G(E)$ in the basic operator expression (6) for the density of states $n(E)$. This Green's-function matrix is the matrix B in expressions (7) or (8), and it is the matrix $\mathcal{G}^h$ in expression (22) and the matrix $\mathcal{G}^{\mathrm{hc}}$ in expressions (52) or (53).

IV. CONCLUSIONS

An expression for the generalized Friedel sum rule has been derived for an arbitrary defect in a dilute alloy, including the as-yet-unexplored defect corresponding to an atom at the saddle-point position in substitutional electromigration. In addition, an expression has been derived that is applicable to an interstitial impurity, which has been used already in the past, but which was only written down previously in analogy to the expression for a substitutional impurity and without explicit derivation. Such expressions are used in practice[7] to ensure or control local charge neutrality in electronic structure calculations for dilute alloys.

It is interesting to note that Lloyd's formula is used, as was done in the past for all similar derivations. Lloyd's formula, however, was written in 1967 for scatterers in free space. The dilute alloy systems studied here would be described more naturally with respect to the unperturbed host as a reference system,[19,27] rather than by using free space as the reference system for both host and dilute alloy.

In a subsequent paper[26] it will be shown that a correspondingly generalized Lloyd formula can indeed be derived.

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APPENDIX A

In this appendix equality (35) and that used in Eq. (51) will be proved. It is a relation between the two host t matrices $\mathcal{T}^h$ and $\mathcal{T}^h_c$, defined by

$$\mathcal{T}^h \equiv (t^{h^{-1}} - B^h)^{-1} , \tag{A1}$$

and, according to (30), by

$$\mathcal{T}^h_c \equiv (t_c^{h^{-1}} - B)^{-1} , \tag{A2}$$

respectively. Definition (A1) can be read from Eq. (23). The subscript c serves to indicate the difference in composition of the matrices t^h in (A1) and t_c^h in (A2). The matrix t^h is composed of all single-site t matrices of the form (4) for the host system. The matrix t_c^h has the same composition as the matrix t^h *outside* the defect region, while all host atoms *inside* the defect region are represented by one t matrix, the host-cluster t matrix T^h_{00} given by (28). Apart from the angular momentum label, which will be omitted throughout, the matrices $\mathcal{T}^h$ and $\mathcal{T}^h_c$ carry position labels. The position label to be used for $\mathcal{T}^h$, indicating all host positions, will be j. The position labels to be used for $\mathcal{T}^h_c$ are 0, indicating the center of the host cluster, and i, indicating all other host atom positions. It will be clear from above that the label i indicates a subset of the range of the label j. Dot labels are used to indicate the host atom positions inside the host cluster, as in (28), which means that the whole set of j labels is given by dot labels and i labels together.

First we prove the following relation between the inverse matrices $\mathcal{T}^{h^{-1}}$ and $\mathcal{T}_c^{h^{-1}}$:

$$\begin{bmatrix} J^{\cdot 0} & 0^{\cdot i''} \\ 0^{i0} & 1^{ii''} \end{bmatrix} (\mathcal{T}_c^{h^{-1}})^{0i'',0i'''} \begin{bmatrix} J^{0\cdot} & 0^{0i'} \\ 0^{i'''\cdot} & 1^{i'''i'} \end{bmatrix} = \mathcal{T}_{jj'}^{h^{-1}} . \tag{A3}$$

Substituting the inverse of (A2) in the left-hand side of (A3), using the notation (28) for the host-cluster t matrix, and carrying out the matrix multiplication, one obtains

$$\begin{bmatrix} J^{\cdot 0} T_{00}^{h^{-1}} J^{0\cdot} & -J^{\cdot 0} B^{0i'} \\ -B^{i0} J^{0\cdot} & (t^{h^{-1}} - B)^{ii'} \end{bmatrix} = \mathcal{T}_{jj'}^{h^{-1}} . \tag{A4}$$

The internal summation over the angular momentum in the nondiagonal terms of the matrix in the left-hand side of (A4), being an unrestricted summation, leads to the equalities

$$\begin{aligned} J^{\cdot 0} B^{0i} &= B^{\cdot i} = B^{h,\cdot i} , \\ B^{i0} J^{0\cdot} &= B^{i\cdot} = B^{h,i\cdot} . \end{aligned} \tag{A5}$$

In the third member the host label h is added, because the B matrices for host position labels are denoted by B^h by definition. The upper-left diagonal elements reduce to $(t^{h^{-1}} - B)^{\cdot\cdot}$, which can be shown by defining

$$F^{0\cdot} \equiv T_{00}^{h^{-1}} J^{0\cdot} . \tag{A6}$$

Using (28), this gives

$$J^{0\cdot} (t^{h^{-1}} - B^h)^{-1} J^{\cdot 0} F^{0\cdot} = J^{0\cdot} . \tag{A7}$$

Multiplying from the left by $J^{\cdot\cdot^{-1}} J^{\cdot 0}$, in which

$$J^{\cdot\cdot} = J^{\cdot 0} J^{0\cdot} \tag{A8}$$

is a positive definite invertible square matrix, one more step leads to

$$J^{\cdot 0}F^{0\cdot}=(t^{h^{-1}}-B^{h})^{\cdot\cdot}\ . \tag{A9}$$

Substituting (A9) with (A6), and (A5) in (A4), one finds

$$\begin{bmatrix}(t^{h^{-1}}-B^{h})^{\cdot\cdot} & -B^{h,\cdot i'}\\ -B^{h,i\cdot} & (t^{h^{-1}}-B^{h})^{ii'}\end{bmatrix}=\mathcal{T}_{jj'}^{h^{-1}}\ , \tag{A10}$$

which is indeed an identity in view of the definition of the position labels, and which establishes (A3).

The required equality is found by first multiplying (A3) from the right by $\mathcal{T}^h$, and from the left by the same matrix occurring as the third factor in the left-hand side of that equation. This leads to

$$\begin{bmatrix}J^{0\cdot}J^{\cdot 0} & 0\\ 0 & 1\end{bmatrix}\mathcal{T}_c^{h^{-1}}\begin{bmatrix}J^{0\cdot} & 0\\ 0 & 1\end{bmatrix}\mathcal{T}^h=\begin{bmatrix}J^{0\cdot} & 0\\ 0 & 1\end{bmatrix}, \tag{A11}$$

in which some position labels are omitted for the sake of surveyability. Multiplying (A11) from the left, first by the inverse of the square matrix

$$\begin{bmatrix}J^{0\cdot}J^{\cdot 0} & 0\\ 0 & 1\end{bmatrix}, \tag{A12}$$

and then by $\mathcal{T}_c^h$, and after that from the right by the same matrix occurring as the first factor in (A3), one finally finds

$$\begin{bmatrix}J^{0\cdot} & 0\\ 0 & 1\end{bmatrix}\mathcal{T}^h\begin{bmatrix}J^{\cdot 0} & 0\\ 0 & 1\end{bmatrix}=\mathcal{T}_c^h\ . \tag{A13}$$

Since in the first equality to be proved only the (00)-matrix element of $\mathcal{T}_c^h$ occurs, (A13) reduces to

$$J^{0\cdot}\mathcal{T}^h_{\cdot\cdot}J^{\cdot 0}=\mathcal{T}^h_{c,00} \tag{A14}$$

being the required relation (35) we are looking for. In the same way (A13) demonstrates the other equality, used in Eq. (51) and mentioned in the lines below it.

These equalities also can be proved in the way they stand, not going via the inverse matrix relation (A3). This might look attractive, but it is not. One has to keep track of the infinite summations in elaborating $\mathcal{T}_c^h$. In fact, it requires diagram techniques to distinguish the i-type summations and the summations inside the host cluster. This could be a nice exercise for a graduate student.

APPENDIX B

In this appendix the following equality will be proven:

$$\operatorname{Im}\operatorname{Tr}\ln J^{0\cdot}A^{\cdot\cdot}J^{\cdot 0}=\operatorname{Im}\operatorname{Tr}\ln A^{\cdot\cdot}\ . \tag{B1}$$

This equality implies the insensitivity of the outcome to the presence of the surrounding matrices $J^{0\cdot}$ and $J^{\cdot 0}$, or, as it is stated in the text, to multiplication of the matrix in the left member from the right with $J^{0\cdot}$, and from the left with $J^{\cdot 0}$. It will be clear that the matrix $J^{\cdot\cdot}$, which shows up that way and which is real and even positively definite, is an irrelevant factor regarding the Im Tr ln operation and therefore can be omitted.

We first give two arguments which may be adequate to convince some readers right away. A first argument is that the matrix manipulation in (B1) is covered by a trace operation, and any trace operation is invariant to a cyclic permutation of the matrices, irrespective of whether they are square or rectangular. Regarding equality (A8) or Eq. (37) without a labels, the desired property would follow immediately. But there may be some doubt because of the presence of the ln operation.

A second argument is an indirect one, and can be derived from the Lloyd formula for the density of states (1) or its integrated form (7). If one treats all scatterers individually, the t matrix defined by (2) has to be substituted in (1). If the scatterers are treated collectively, as a cluster, the t matrix defined by (27) has to be substituted. The outcome, being the (integrated) density of states of the system, cannot be different, and since the internal matrix $T^{\cdot}$ in (27) is precisely the matrix defined by (2), equality (B1) is proven implicitly by this.

Equality (B1) is proven most simply in its differential form, from which it originates and by which one gets rid of the ln operation. The derivative of the left-hand side of (B1) with respect to the energy can be written as

$$\begin{aligned}\frac{d}{dE}\operatorname{Im}\operatorname{Tr}\ln J^{0\cdot}A^{\cdot\cdot}J^{\cdot 0}&=\operatorname{Im}\operatorname{Tr}\left\{\frac{1}{J^{0\cdot}A^{\cdot\cdot}J^{\cdot 0}}\frac{d}{dE}J^{0\cdot}A^{\cdot\cdot}J^{\cdot 0}\right\}\\ &=\operatorname{Im}\operatorname{Tr}\left\{\frac{1}{J^{0\cdot}A^{\cdot\cdot}J^{\cdot 0}}\left[\frac{dJ^{0\cdot}}{dE}A^{\cdot\cdot}J^{\cdot 0}+J^{0\cdot}A^{\cdot\cdot}\frac{dJ^{\cdot 0}}{dE}+J^{0\cdot}\frac{dA^{\cdot\cdot}}{dE}J^{\cdot 0}\right]\right\}\ .\end{aligned} \tag{B2}$$

Using the method as outlined in Sec. (III A) and represented by equalities (36) and (42), one finds that

$$J^{\cdot 0}\frac{1}{J^{0\cdot}A^{\cdot\cdot}J^{\cdot 0}}J^{0\cdot}=A^{\cdot\cdot^{-1}}\ . \tag{B3}$$

Applying this equality to the third term, this term becomes precisely the energy derivative of the right-hand side of (B1). Applying this equality to the first and second terms in (B2), the matrix $A^{\cdot\cdot}$ appears to cancel, and these terms obtain the form

$$\operatorname{Im}\operatorname{Tr}\left\{\left[J^{\cdot 0}\frac{dJ^{0\cdot}}{dE}+\frac{dJ^{\cdot 0}}{dE}J^{0\cdot}\right]\Big/J^{\cdot\cdot}\right\}=\operatorname{Im}\operatorname{Tr}\left\{\frac{dJ^{\cdot 0}J^{0\cdot}}{dE}\Big/J^{\cdot\cdot}\right\}=\frac{d}{dE}\operatorname{Im}\operatorname{Tr}\ln J^{\cdot\cdot}\ , \tag{B4}$$

which evidently is zero. By this the equality (B1) has been established in its differential form, but due to the explicit presence of the derivative with respect to the energy it also holds in the form as it stands.

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