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Formation volume and energy of a vacancy: lattice distortion and non-linear screening in aluminium

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Abstract. The contribution due to non-linear (third order) screening to the amplitude of asymptotic lattice distortion around a vacancy is found to be not less than 30–60%, depending on the approximation for dielectric screening. Though also important in principle, the fourth order term is much smaller. The calculated values for the energy and volume of formation are reasonable, and a peculiar shape for the elastic diffuse neutron scattering pattern is predicted.

Introduction

The electron liquid-pseudopotential method seems to be a most adequate approach to calculate the *distortion* of the lattice due to a defect and, in particular, to a vacancy [1, 2, 3, 4] in simple metals. Besides giving reasonably good numerical results, the main virtue of the method lies in the straightforward way by which the total energy of the metal is obtained as an explicit function of the ionic coordinates, including both the pairwise interactions and the many-ion contributions. Having at hand such an expression, the determination of the equilibrium configuration of the ions and the dynamics of the lattice is also straightforward. Using this method, however, some intriguing problems may arise, like the following: a substantial difference has been found [4] between two formulae, each giving the volume of formation for a vacancy, calculated either by

- (i) the rigid pair force model of lattice statics or
- (ii) applying the zero pressure condition to the total energy of the crystal.

Though method (ii) was shown [4] to be the correct one to obtain the formation volume, it has not been clear if the lattice statics method, when combined with the pseudopotential approach, is inherently wrong, or if not, which way should it be completed. Subsequently, the analogous problem of the volume change and, in general, the lattice distortion has been studied for dilute substitutional alloys [5, 6] and it has been demonstrated that in all cases the lattice statics method *does*, in fact, give the same result as method (ii), provided the forces related to non-linear screening (many-ion forces) are also taken into account. Now, since the real metal can be considered as a heterovalent substitutional solution, with vacancies as solute 'atoms', the formulae given for a heterovalent alloy [6] directly apply. It

follows [5, 6] that the consistent lattice statics calculation accurate up to the order $(v_{\mathbf{G}}/E_F)^2$ must include the third and fourth order screening functions [7, 8] in calculating the distorting forces. In view of the complicated form of these functions, however, the numerical calculation can be done in the following way: (a) the formation volume and, thereby, the displacements in the asymptotically far region are calculated by method (ii), and (b) the displacements of the neighbouring atoms are determined by the linear screening terms only, though being aware of the fact that these lattice statics results will not join, in the far region, to the correct asymptotic solution, as given by (a). (Here $v_{\mathbf{G}}$ is the Fourier component of the bare ion-electron potential at a reciprocal lattice vector \mathbf{G} and E_F is the Fermi energy.) In this way, the higher order screening contributions can be determined at $\mathbf{q} \rightarrow 0$. The aim of the present work is

- (a) to study the third and fourth order contributions to the asymptotic displacement amplitude separately, and
- (b) to calculate the displacement field in real and momentum space and to compare this with the results of the elasticity theory. In doing that we arrive at predicting an unusual diffraction pattern to be expected from neutron scattering by frozen vacancies in aluminium.

Basic formulae

For the formation volume Ω_f the zero pressure condition [5, 6, 9] leads in a straightforward way to

$$\left(\frac{\Omega_f}{\Omega_0} - 1\right) \equiv \frac{\delta\Omega}{\Omega_0} = -\frac{1}{B} \frac{d(\Delta E)}{d\Omega_0} \quad (1)$$

where B is the bulk modulus and ΔE is the derivative of the total energy with respect to vacancy concentration

$$\Delta E = \left(\frac{dE_{\text{tot}}}{dc}\right)_{c=0} \quad (2)$$

and Ω_0 is the volume per atom in the pure metal. The displacement \mathbf{u} of the \mathbf{l} th ion in the lattice is given by [9]

$$u(\mathbf{l})_{\alpha} = \sum_{\mathbf{l}'} g(\mathbf{l}-\mathbf{l}')_{\alpha\beta} f(\mathbf{l}')_{\beta} \quad (3)$$

where g is the static response function (the inverse of the dynamical matrix of the lattice) and \mathbf{f} is the displacing force acting upon an ion due to the presence of the defect at $\mathbf{l}=0$. When taking into account only pairwise interionic forces, for the case of a vacancy \mathbf{f} is obviously the negative of the pair force; generally it contains also multi-ion contributions.

For a simple metal where the perturbed electron liquid formalism is expected to work, all quantities in the right hand side of (1) and (3) can be expressed as a series in terms of the bare ion-electron potential v of a single ion and the dielectric functions $\epsilon(\mathbf{q})$, $\epsilon(\mathbf{q}, \mathbf{q}')$... of the homogeneous electron gas [6-8]. The expression for g is standard [5, 9], while for ΔE and \mathbf{f} the formulae of Ref. [6] apply with taking the 'solute atom potential' equal to zero. To 2nd order in $v_{\mathbf{G}}$ we

obtain

$$\Delta E = -Z \left(\varepsilon_0 - \frac{r_s}{3} \frac{d\varepsilon_0}{dr_s} \right) + \frac{Z^2 e^2}{R_a} \gamma - \frac{2Z\beta}{\Omega_0} + \sum_{\mathbf{G}} |V_{\mathbf{G}}|^2 \phi_{\mathbf{G}} + Z \cdot \frac{1}{2} \sum_{\mathbf{G}} |v_{\mathbf{G}}|^2 \frac{\partial \phi_{\mathbf{G}}}{\partial \eta_0} - \frac{1}{2} \frac{\Omega_0}{(2\pi)^3} \int d\mathbf{q} |v_{\mathbf{q}}|^2 \phi_{\mathbf{q}} + E_{\text{rel}} \quad (4)$$

where $Z = 3$ for Al, R_a is the radius of the Wigner-Seitz sphere of volume Ω_0 , r_s is that for one electron, ε_0 is the energy per electron in the homogeneous liquid of density n_0 , and γ is the Madelung constant for the (fcc) lattice. The function ϕ is related to the dielectric function as

$$\phi(\mathbf{q}, n_0) = \frac{q^2}{4\pi e^2} \left(1 - \frac{1}{\varepsilon(\mathbf{q}, n_0)} \right) \quad (5)$$

The constant β is determined by the average ion electron potential as

$$\beta = \int \left(v(\mathbf{r}) + \frac{Ze^2}{|\mathbf{r}|} \right) d\mathbf{r} \quad (6)$$

and E_{rel} is the energy liberated by the relaxation of the lattice [9],

$$E_{\text{rel}} = -\frac{1}{2} \sum_{\mathbf{l}} \mathbf{f}(\mathbf{l}) \cdot \mathbf{u}(\mathbf{l}) \quad (7)$$

with \mathbf{u} 's taken from (3).

Formulae (1)–(7), with the linear screening approximation in (3), have previously been used, though in a different form, to calculate vacancy characteristics [1–4]. Here we analyze the nonlinear screening parts in (3) for large $|\mathbf{l}|$, and also calculate the displacement field in \mathbf{q} -space, with reference to the expected diffraction picture.

For the force we have

$$\mathbf{f}(\mathbf{l}) = \boldsymbol{\varphi}(\mathbf{l}) - \frac{i}{N} \sum_{\mathbf{k}} \mathbf{k} F(\mathbf{k}) e^{i\mathbf{k}\mathbf{l}} \quad (8)$$

with

$$\boldsymbol{\varphi}(\mathbf{l}) = Z^2 e^2 \frac{\partial}{\partial \mathbf{r}} \left\{ \frac{1}{r} \text{erfc}(\eta r) \right\}_{\mathbf{r}=\mathbf{l}} \quad (9)$$

and

$$F(\mathbf{k}) = -Z^2 e^2 \frac{4\pi}{\Omega_0 k^2} e^{-k^2/4\eta^2} + \sum_{m=2}^{\infty} F^{(m)}(\mathbf{k}) \quad (10)$$

where η is the arbitrary constant of an Ewald transformation, and the sum represents the contribution of the electron polarization of different orders. For $m=2$ we have the electron component of the interionic pair potential, with a minus sign,

$$F^{(2)}(\mathbf{k}) = |v_{\mathbf{k}}|^2 \phi_{\mathbf{k}} \quad (11)$$

which is the linear screening contribution. Generally, $F^{(m)}$ ($m > 2$) contains m

factors of $v_{\mathbf{k}}$ and the (nonlinear) response function of m -th order, with $(m-1)$ variables in momentum space [6].

By (3) we arrive at the asymptotical displacements^{5,6,10} as

$$\mathbf{u}(\mathbf{l}) \sim \Omega_0 \frac{\mathbf{A}(\mathbf{l})}{|\mathbf{l}|^2} \cdot \frac{\delta\Omega}{\Omega_0} \quad (12)$$

with

$$\frac{\delta\Omega}{\Omega_0} = \frac{1}{B\Omega_0} \lim_{\mathbf{q} \rightarrow 0} \left\{ F(\mathbf{q}) + \sum_{\mathbf{G} \neq 0} \left[F_{\mathbf{q}+\mathbf{G}} + \frac{1}{3} G_{\gamma} \frac{\partial F_{\mathbf{q}+\mathbf{G}}}{\partial G_{\gamma}} \right] - \frac{1}{3} \sum_{\mathbf{l}} \varphi(\mathbf{l})_{\gamma} l_{\gamma} \right\} \quad (13)$$

Here the anisotropy amplitude \mathbf{A} is deduced *uniquely* from the inverse Christoffel matrix of the metal, thus it is a given function of the elastic constants [5, 10]. The formula (13) is valid in the approximation

$$\left| \frac{df_i}{dl_k} \right| |\mathbf{u}| \ll |\mathbf{f}| \quad (14)$$

which is taken to be granted in this case. The contributions from $F^{(m)}$ to the sum in (13) is easy to calculate, without making explicit use of their form at a general argument [6–8]. For example, there appears

$$F^{(4)}(\mathbf{q} \rightarrow 0) = Z \sum_{\mathbf{G} \neq 0} \frac{1}{2} |V_{\mathbf{G}}|^2 n_0 \frac{\partial^2 \phi(\mathbf{G}, n_0)}{\partial n_0^2} \quad (15)$$

and terms of similar structure [6], containing the operation $\partial/\partial n_0$, related to $F^{(3)}(0)$ and $F^{(3)}(\mathbf{G})$. It is by taking into account these terms that (12)–(13) and (1)–(2) become equivalent.¹⁾

We proceed now as follows: taking a particular form for the ion-electron potential v , used already with success to explain the properties of the dilute **Al**–**Li** system, we calculate Ω_f by (2) and (4), and study the weight of terms corresponding to $F^{(3)}$ and $F^{(4)}$ in the result. The results for Ω_f , as well as those for the energy of formation

$$\begin{aligned} E_f = \Delta E + E_{id} = & Z \frac{r_s}{3} \frac{d\varepsilon_0}{dr_s} + \frac{1}{2} \frac{Z^2 e^2}{R_a} \gamma - \frac{Z\beta}{\Omega_0} \\ & + \frac{1}{2} \sum_{\mathbf{G} \neq 0} |v_{\mathbf{G}}|^2 \phi_{\mathbf{G}} - \frac{1}{2} \frac{\Omega_0}{(2\pi)^3} \int d\mathbf{q} |v_{\mathbf{q}}|^2 \phi_{\mathbf{q}} + \\ & + Z \frac{1}{2} \sum_{\mathbf{G} \neq 0} |v_{\mathbf{G}}|^2 \frac{\partial \phi_{\mathbf{G}}}{\partial n_0} + E_{rel} \end{aligned} \quad (16)$$

and for the displacement field $\mathbf{u}(\mathbf{l})$ are analyzed in the next section (here E_{id} is the energy per atom in the pure metal, representing the energy of the atom removed to the surface).

¹⁾ In (13) terms corresponding to $dE_{rel}/d\Omega_0$ (cf. (1)–(4)) are lacking, as a result of the approximation (14); this term will indeed be shown negligible.

Numerical results

In the numerical calculations a local ion-electron potential of the Heine–Abarenkov type

$$v(\mathbf{r}) = \begin{cases} -\frac{Ze^2}{|\mathbf{r}|} & |\mathbf{r}| > R_c \\ -\lambda \frac{Ze^2}{R_c} & |\mathbf{r}| \leq R_c \end{cases} \quad (17)$$

was used, of the same form as in previous work [1–4, 6, 8, 9], with the parameters R_c , λ determined [6] by fitting to the observed volume and phonon spectrum of pure aluminum: $R_c = 1.27$ a.u. and $\lambda = 0.332$ (Toigo–Woodruff screening function [11]) and, alternatively, $R_c = 1.25$ a.u. and $\lambda = 0.361$ (Geldart–Vosko screening function [12]).

The results for the volume and energy of formation are shown in Tables I and II. If one proceeds by calculating the volume of formation via the asymptotic

Table I

Contributions to the asymptotic amplitude $\delta\Omega/\Omega_0$ from linear, third and fourth order polarization (equations (12)–(13)). The result neglecting the pressure from E_{rel} is in the 4th column, while the inclusion of this gives the value in column 5. The notation GV and TW refers to different approximations for the dielectric screening (see text), $\Omega_f = \Omega_0 + \delta\Omega$.

	$\delta\Omega/\Omega_0$					
	$F^{(2)}$	$F^{(3)}$	$F^{(4)}$	$\left(\frac{\Omega_f}{\Omega_0}\right)_{\text{n.d.}}$	$\frac{\Omega_f}{\Omega_0}$	$\left(\frac{\Omega_f}{\Omega_0}\right)_{\text{exp}}$
GV	–0.11	–0.19	–0.01	0.69	0.67	0.62 ^a
TW	–0.14	–0.06	–0.02	0.78	0.75	
previous work					0.68 ⁽⁴⁾	

^a) R. M. Emrick and P. B. McArdle, Phys. Rev. 188, 1156 (1969).

Table II

Contributions to the energy of formation of each term in equation (16) in ry units. The notation GV and TW is the same as in Table I.

	Hom. el. liqu.	Madel.	av.	"inc"	"coh"	$\frac{\partial}{\partial n} ()$	relax.	E_f	$(E_f)_{\text{exp}}$
GV	–0.560	5.411	–1.205	–3.676	0.045	0.042	–0.001	0.055	0.049 ^(a)
TW			–1.278	–3.645	0.054	0.051	–0.002	0.031	

(a) A. Seeger, J. Phys. F 3, 248 (1973).

amplitude of the displacements, equations (12)–(13), one gets the result shown in Table I, where the 1st column is the value for the linear screening approximation, and the next two columns contain the third and fourth order contributions.²⁾ The sum of these three numbers gives the ‘non-distorted’ value for $\partial\Omega/\Omega_0$, so that

²⁾ The TW screening function does not satisfy the ‘compressibility limit’ [8] exactly, so that its limiting value at $\mathbf{k} \rightarrow 0$ is slightly in error. This is corrected for in the 1st column of Table 1.

the part originating in the pressure associated with the relaxation energy (cf. (1), (4)), has still to be added, with the result of column 5. One sees that

- (i) the contribution of $F^{(3)}$ is of the same order as the linear screening (or 'rigid pair force') result;
- (ii) the volume change coming from the derivative of the energy of relaxation is by an order of magnitude smaller, and the same is true for the contribution of $F^{(4)}$.

Neglecting here $F^{(3)}$ besides the terms coming from $F^{(2)}$ would thus have no sense, and the near cancellation of $F^{(3)}$ and $F^{(4)}$ at some particular potential parameters is indeed fortuitous [4]. On the other hand, the volume change associated with the 'pressure' of E_{rel} is only 2–3%, in agreement with previous work [4], hence the terms (left out by the approximation (14)) are really small.

The interionic potential in Al corresponding to (17) with the chosen values of R_c and λ is shown in Fig. 1. As pointed out before, its (positive) gradient is precisely the distorting force $\mathbf{f}(\mathbf{l})$ around a vacancy (Fig. 2). Remarkable is the change in the potential and the force on changing the approximation for the screening function. We note the similarity of our interionic potential for the *TW* screening with the one obtained by the authors of Ref. [13], with the main feature of *not being negative* near its first minimum. The calculated radial displacements in real space are shown in Fig. 3, as compared to that expected for an isotropic continuum for which equation (12) applies *everywhere* with $\mathbf{A} = \hat{\mathbf{l}} \cdot \mathbf{B}/4\pi C_{11} \approx 0.055\hat{\mathbf{l}}$. The lattice calculation is done for forces corresponding to the linear screening approximation (11), the dashed curve is for the continuum with $\Omega_f/\Omega_0 = 0.78$. Asymptotically, the points should approach the curve (in fact that for $\delta\Omega/\Omega_0 = 0.86$, according to Table I, corresponding to the neglect of $F^{(3)}$ and $F^{(4)}$), but this 'far' region seems not to be attained even for the distant neighbours like those at (880).

The fact that the elastic continuum approximation is completely inadequate in our case is, however, much better seen in wavenumber space, Fig. 4. The

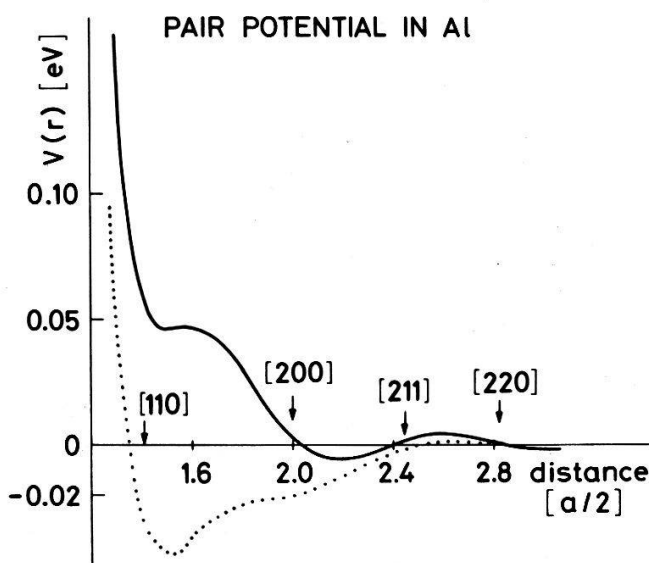


Figure 1

The pairwise interaction in Al metal calculated from the 2-parameter potential, equation (17). The two curves correspond to the *TW* (solid line) and *GV* (dotted line) screening functions; the arrows indicate the positions of near neighbors.

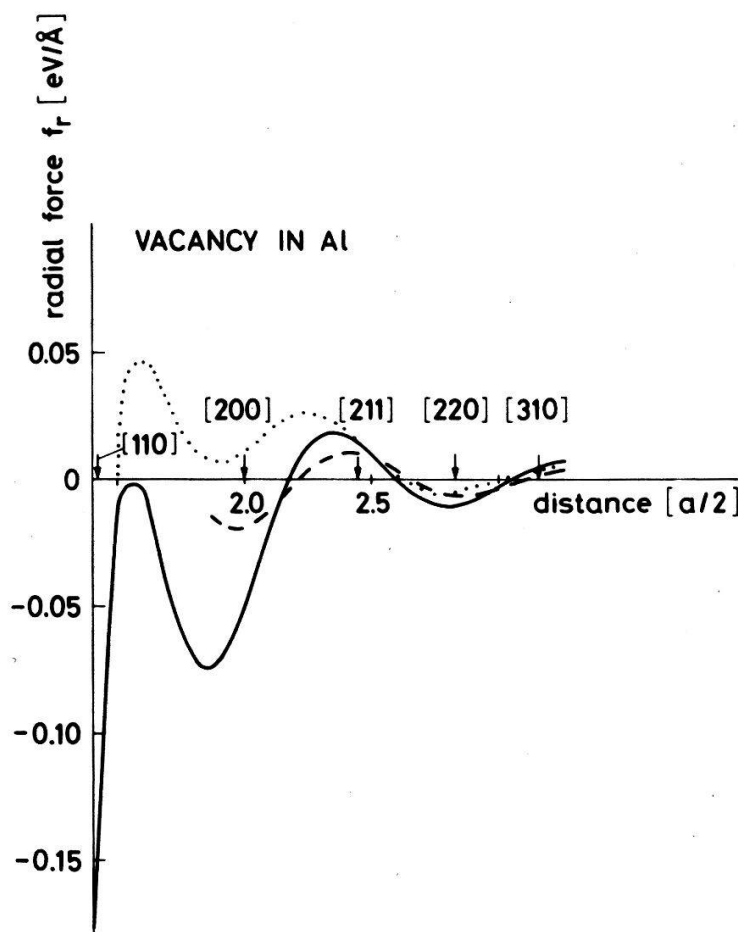


Figure 2

The (radial) distorting force near a vacancy in Al, as derived from the potentials of Fig. 1. The asymptotic behaviour with a shift in phase is shown by the dash-dotted line.

displacements in an elastic continuum would follow equation (12) *everywhere*, thus $i|q||u_q|$ would be a straight line parallel to the q axis at $(B/\tilde{c})(\Omega_f/\Omega_0 - 1)$ (\tilde{c} is a linear combination of the elastic constants such [6] that the longitudinal sound velocity at the direction \hat{q} is $\tilde{c}_l(\hat{q}) = (\tilde{c}\Omega_0/M)^{1/2}$). If we look now [15], instead of a vacancy, at the displacement field near a solute Ga atom in Al, where the substitutional Ga^{3+} ions perturb surely much less the host lattice than a vacancy, we obtain the dash-dotted curve. Though even this is not a horizontal straight line (*any* theory taking into account the discrete lattice structure and symmetry *must* result at a vanishing of u_q at half of a reciprocal lattice vector [6, 16]), the curve for the Ga impurity is quite structureless and is 'as near as possible' to the predictions of a continuous theory. This is strikingly *not* the case for the vacancy as defect, since *two additional nodes of u_q appear* and the function has a rather large slope at $q = \frac{1}{2}G$. Similar form for u_q appears also for a solute Li atom [6] which has actually been observed as a hump and two valleys in the neutron scattering pattern.³⁾

Remind now that the curves in Fig. 4 have been calculated by neglecting nonlinear screening terms, so that their values at $|q| \rightarrow 0$ must be in error, as discussed above. On the base of Table I we can, in fact, predict the *correct* values

³⁾ The coherent diffuse neutron cross section is, to a good approximation [6, 15], proportional to iqu_q .

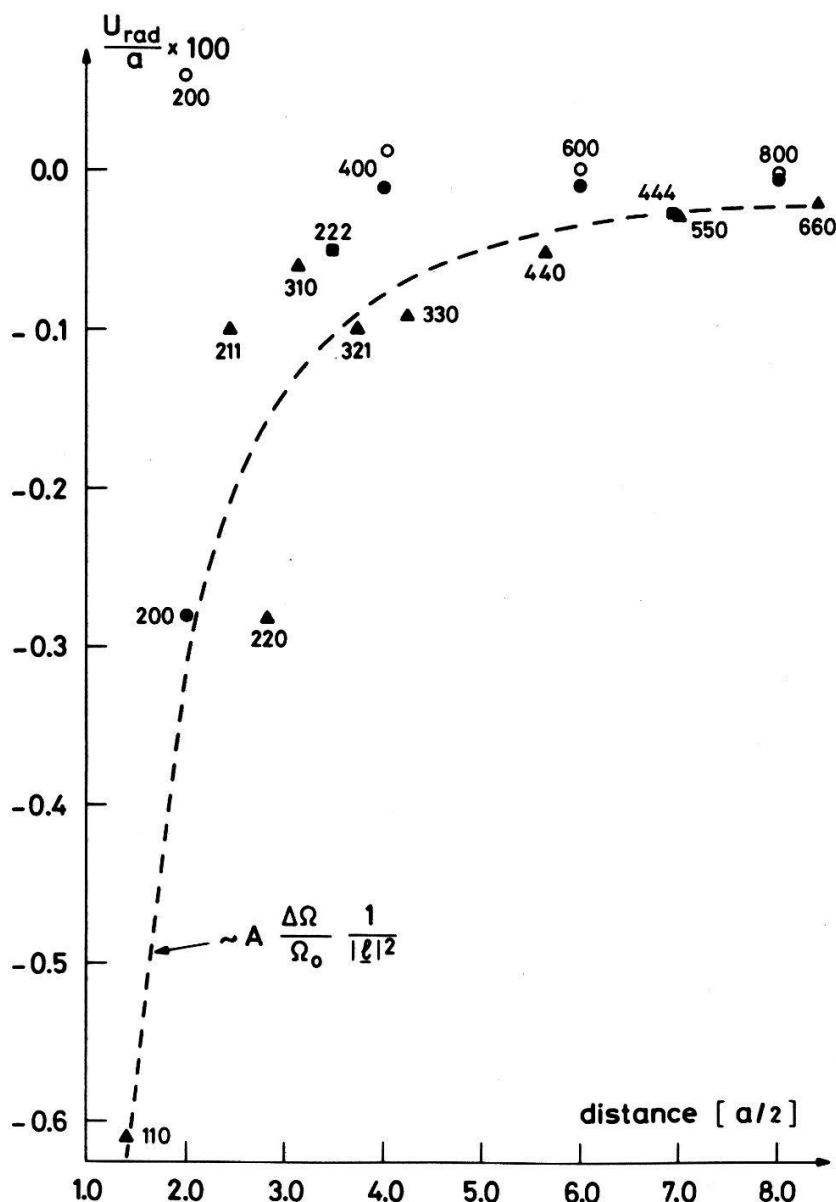


Figure 3

Predicted radial displacements of the Al atoms near a vacancy (●, ▲, ■ : TW, ○ : GV). The dashed line is the isotropic continuum approximation.

by using column 4. We get for $i\mathbf{q}\mathbf{u}_q$ the value -0.15 for the TW dielectric function and -0.20 (!) for the GV one, which points clearly to the great role of $F^{(3)}$ and $F^{(4)}$ in the diffraction pattern.

The results for each of the 7 contributions to E_f , at the right hand side of equation (16), are shown in Table II.

We see that the contribution from the homogeneous electron gas, the Madelung energy, the average potential term and the 'incoherent' ($\int d\mathbf{q}$) terms, each of them being of the order of 1 ry or more, strongly cancel, so that the result for the formation energy is less than 0.1 ry. The cancellation appears also in the calculation of the volume change, but there only the first three terms are competitive. The energy of relaxation itself is very small, even compared to the formation energy.

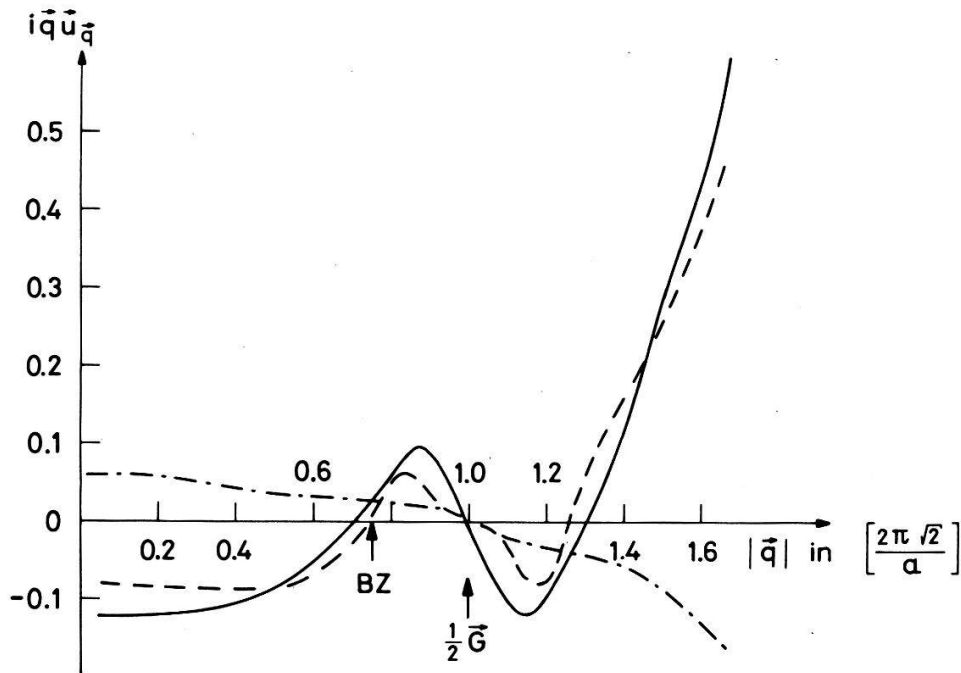


Figure 4

The displacement field in q -space, approximately proportional to the neutron scattering cross section [16]. Besides the result for the vacancy (full line : TW, dashed line : GV) that expected around a Ga impurity in Al is also plotted (dash-dotted line). The curves, as calculated in linear screening approximation, tend to a limit at $q \rightarrow 0$ which is not the correct one (-0.15 and -0.20 , respectively, for TW or GV screening; see text).

In conclusion we can say that

- (i) The error in calculating the amplitude of the long range displacement field at a vacancy with retaining only the linear screening term in lattice statics is quite large for both screening approximations, leading to an amplitude of 30 or 60% smaller than the correct one. The essential nonlinear term proves to be $F^{(3)}$, representing 3-ion interactions. It is this amplitude $\delta\Omega/\Omega_0$ which determines the diffuse elastic neutron scattering cross section at $q \rightarrow 0$, so that the effect of $F^{(3)}$ in the scattering pattern all over the range $|\mathbf{q}| \ll |\mathbf{G}|$ is *accordingly large*.
- (ii) The expected displacement field *does not resemble at all* the behaviour of an elastic continuum and has a shape fairly similar to that measured for a Li impurity in aluminum. This is reasonable, since the change in ionic charge $Z_{\text{imp}} - Z_{\text{host}}$ equals -2 for Li and -3 for a vacancy.
- (iii) The contribution of the relaxation energy to both the energy and the volume of formation is very small, less than 10%, in agreement with previous work.

The experimental neutron scattering study of the deformation field near a mono-vacancy would be of great value.

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