Electric field gradient in dilute alloys of aluminum

S. D. Raj

Department of Physics, Panjab University, Chandigarh-160014, India

J. Singh

Department of Physics, Panjab Agricultural University, Ludhiana, 141004, India

S. Prakash

Department of Physics, Panjab University, Chandigarh-160014, India

The electric field gradient q and asymmetry parameter η are calculated for AlMg, AlSi, AlCu, AlZn, AlGa, AlGe, AlAg, and AlCd alloys. The Alfred and Van Ostenburg charge perturbation, which accounts for both the asymptotic and preasymptotic regions, is used to evaluate the valence-effect contribution. The size-effect contribution is calculated in the elastic continuum model proposed by Sagalyn and Alexander. It is found that the valence effect alone cannot explain the observed q and η. The size-effect contribution is found larger than the valence-effect contribution for all the alloys except for AlAg for which the valence-effect contribution is found larger than the size-effect contribution. The calculated values of q and η are found to be in close agreement with the experimental values and exhibit an improvement over other available theoretical results.

I. INTRODUCTION

In metallic alloys with cubic symmetry, as dilute alloys of Cu and Al, the substitutional impurity produces a charge perturbation with lower symmetry that gives rise to an electric field gradient (EFG) q at nearest-neighbor nuclei. The contribution due to valence and the size differences between the impurity and the host atoms are called the valence and size effects, respectively. These effects are correlated and contribute towards the EFG.[1−5] We discuss here the q and η in the dilute alloys of Al for nontransition-metal impurities. Al behaves as a free-electron metal in many respects[6] and therefore one expects that the Kohn and Vosko[1] and Blandin and Friedel[2] theory for EFG should be fairly applicable.

Very few attempts[7−9] have been made to explain the observed values of EFG and asymmetry parameter η in dilute alloys of Al. Nevald et al.[8] calculated the EFG in the free-electron model obtaining the phase shifts from the scattering potential adopting the Alfred and Van Ostenburg[10] scheme. Fukai and Watanabe[7] used pseudopotential theory to calculate the EFG and wipe-out numbers. Jena et al.[11] incorporated the effect of nonlinear screening in the evaluation of EFG for dilute alloys of Al. However, all these authors considered only the valence-effect contribution which exhibits perfect cylindrical symmetry thereby reducing η to zero contrary

to experimental observations.[12,13] Furthermore, their calculated values of EFG are uneven with the experimental results. With the use of the Sagalyn and Alexander[5] approach for calculating the valence- and size-effect contributions, Prasad Rao and Mohapatra[9] calculated the EFG and η for AlMg alloy and further extended their calculations for the Al-vacancy system. Recently, Ponnambalam and Jena[14] developed a formalism for the EFG due to a point defect in a cubic metal where the charge perturbation is calculated self-consistently in the density-functional approach and the size effect is calculated using an oscillatory atomic displacement field. These authors explained the EFG and η for a monovacancy in Al up to the fourth-nearest neighbors. To the best of our knowledge, the EFG and η are not yet studied for most of the Al-based alloys and therefore needs a systematic investigation.

In our earlier paper[15] (hereafter referred to as paper I) the valence-effect contribution is evaluated using the preasymptotic charge perturbation due to Alfred and Van Ostenburg[10] in the Kohn and Vosko[1] approach. The size effect as suggested by Sagalyn and Alexander[5] is added to calculate the EFG and η for Cu-based alloys and reasonably good agreement with experimental results is obtained. Therefore, it is interesting to extend the calculations for Al-based alloys also. However, we restrict our calculations to nontransitional impurities,

as these do not form bound and virtual-bound states.[16]

This paper is organized as follows: Section II contains the necessary theory in brief. The results are presented and discussed in Sec. III. Conclusions are drawn in Sec. IV.

II. THEORY

The general expression for the valence-effect contribution to EFG along the line joining the impurity and the host atom (assumed as Z axis) is given as[1,2]

$$q_{\parallel}^{V}(\vec{r}_n) = q_{ZZ}^{V}(\vec{r}_n)$$

$$= -\int \frac{\Delta n(\vec{r}+\vec{r}_n)}{r^3}[1+\gamma(r)]$$

$$\times (3\cos^2\theta - 1)d\vec{r} . \qquad (1)$$

Here $\gamma(r)$ is the Sternheimer antishielding factor, $\vec{r}_n$ is the position of the nth-nearest neighbor from the impurity, and $\Delta n(\vec{r})$ is the charge perturbation produced by the impurity. Kohn and Vosko[1] and Blandin and Friedel[2] derived an analytical expression for the EFG tensor using an asymptotic expression of the free-electron charge perturbation. Alfred and Van Ostenburg[10] improved the expression by including one more term, known as the preasymptotic contribution. With the use of the Alfred and Van Ostenburg[10] free-electron charge perturbation in Eq. (1) we get the analytical expression for the valence-effect contribution[15] as

$$q_{\parallel}^{V}(\vec{r}_n) = q_{ZZ}^{V} = \frac{8\pi}{3}\alpha \left[A \frac{\cos(2R_F r_n + \phi)}{r_n^3} \right.$$

$$+ B \frac{\cos(2k_F r_n + \xi)}{r_n^4}$$

$$\left. - \frac{3}{2} \frac{A\sin(2k_F r_n + \phi)}{k_F r_n^4} \right] , \qquad (2)$$

where α is the core-enhancement factor. The constants A, B, ϕ, and ξ are related to the phase shifts $\eta_l(\vec{k})$ and their derivatives $[\partial\eta_l(\vec{k})/\partial k]$ at $k=k_F$, the Fermi momentum by the relations

$$A\cos\phi = -\frac{1}{4\pi^2}\sum_l (2l+1)(-1)^l \sin 2\eta_l^{k_F} ,$$

$$A\sin\phi = -\frac{1}{2\pi^2}\sum_l (2l+1)(-1)^l \sin^2\eta_l^{k_F} , \qquad (3)$$

$$B\cos\xi = \frac{1}{4\pi^2}\sum_l (2l+1)(-1)^l$$

$$\times \left[\frac{2l(l+1)}{k_F}\sin^2\eta_l^{k_F} \right.$$

$$\left. + \left[\frac{\partial\eta_l}{\partial k} \right]_{k_F} \sin 2\eta_l^{k_F} \right] ,$$

$$B\sin\xi = -\frac{1}{4\pi^2}\sum_l (2l+1)(-1)^l$$

$$\times \left[\frac{l(l+1)}{k_F}\sin 2\eta_l^{k_F} \right.$$

$$\left. + \left[\frac{\partial\eta_l}{\partial k} \right]_{k_F} \cos 2\eta_l^{k_F} \right] .$$

The valence-effect contribution exhibits cylindrical symmetry about the Z axis, therefore the components of the EFG perpendicular to $\vec{r}_n$ ($q_{\perp}^{V}$) and along the x direction (q_{xx}^{V}) are given as

$$q_{\perp}^{V} = q_{xx}^{V} = -\tfrac{1}{2}q_{\parallel}^{V} . \qquad (4)$$

Here x is the crystal coordinate system.

There are three effects that result from the difference in the size of the host and impurity atoms. First is the indirect size effect which has been taken care of by evaluating the EFG at the displaced atomic positions appropriate to an alloy assuming the displacement to be spherically symmetric as

$$\vec{u} = \frac{D}{r^2}\frac{\vec{r}}{r} , \qquad (5)$$

where

$$D = \left[\frac{3}{16\pi\gamma_E} \right]a^3 \left[a^{-1}\frac{da}{dc} \right] .$$

γ_E is a function of Poisson ratio and $a^{-1}(da/dc)$ is the fractional change in lattice parameter per atomic percent concentration.

Second is also an indirect size effect that is included by using the modified Friedel sum rule, defined as

$$Z_{eff} = Z_i - Z_h \left[1 + \frac{3}{\gamma_E}\frac{1}{a}\frac{da}{dc} \right]$$

$$= \frac{2}{\pi}\sum_l (2l+1)\eta_l^{k_F} , \qquad (6)$$

in evaluating the phase shifts. Z_i and Z_h are the valencies of the impurity and the host atoms,

respectively.

Third is the direct size effect and is formulated by Sagalyn and Alexander[5] assuming a spherically symmetric displacement[4] of the form given in Eq. (5). We use the same expressions as given by Sagalyn and Alexander. The components of the EFG at first-nearest neighbors (1NN) are

$$q_{\parallel}^S = -\tfrac{1}{3}q_{\perp}^S = \tfrac{1}{2}q_{xx}^S = \frac{27\sqrt{2}}{16\pi\gamma_E}\left[\frac{1}{a}\frac{da}{dc}\right]\frac{\lambda}{r_1^3} ,$$

(7)

and at second-nearest neighbors (2NN) they are

$$q_{\parallel}^S = -2q_{xx}^S = -2q_{yy}^S = \frac{108\sqrt{2}}{16\pi\gamma_E}\left[\frac{1}{a}\frac{da}{dc}\right]\frac{\lambda}{r_2^3} .$$

(8)

Here λ is the size-effect parameter that relates the experimental EFG to the EFG that would be observed if the distorted lattice is made up of unshielded point charges.

We assume the EFG coordinate system as (X,Y,Z). Here the valence-effect contribution is evaluated in EFG coordinates (X,Y,Z) and size-effect contributions are evaluated in the crystal coordinate system (x,y,z). These two contributions are combined in the same manner as in paper I to obtain the components of total EFG, q_{XX}, q_{YY}, and q_{ZZ}. The asymmetry parameter η is defined as

$$\eta = \frac{q_{XX} - q_{YY}}{q_{ZZ}} ,$$

(9)

where it is assumed that $|q_{XX}| \le |q_{YY}| \le |q_{ZZ}|$.

III. RESULTS AND DISCUSSIONS

We are investigating the EFG for simple metal impurities in Al. Therefore, it is assumed that only s and p phase shifts, i.e., η_0 and η_1 will be significant and higher-order phase shifts will be negligible.[16,17] η_0 and η_1 are determined by satisfying the Friedel sum rule, given by Eq. (6), and the experimental residual resistivity given as

$$\Delta\rho = \frac{4\pi}{Z_h k_F}\sum_l (l+1)\sin^2(\eta_l - \eta_{l+1}) .$$

(10)

We find different sets of η_0 and η_1 that satisfy Eqs. (6) and (10) simultaneously. Out of these sets we choose a set in which phase shifts decrease rapidly. $(\partial\eta_l/\partial k)_{k_F}$ are calculated in the Alfred and Van Ostenburg's approximation,[10] i.e.,

$$\eta_l(k) \simeq \alpha_l k^{2l+1} ,$$

(11)

where α_l is a constant.

Equation (5) is used to evaluate the displaced position of the nth-nearest neighbor r_n of the impurity given as

$$r_n = \sqrt{n}\,d_0 + \frac{3}{4\pi\gamma_E}\frac{\Omega_0}{(\sqrt{n}\,d_0)^2}\left[\frac{1}{a}\frac{da}{dc}\right] ,$$

(12)

where d_0 is the distance of 1NN and Ω_0 is the atomic volume of the host lattice. The phase shifts, their derivatives calculated with the help of Eq. (11), the distances of 1NN and 2NN given by Eq. (12), $Z_{\rm eff}$ given by Eq. (6), and other physical parameters for Al alloys are given in Table I. The calculated values of A, B, ϕ, and ξ are also tabulated there.

The core-enhancement α has been the most uncertain parameter in evaluating the valence-effect contribution. Blandin and Friedel[2] calculated $\alpha = 10$ for Al using a single-orthogonalized plane-wave theory. Fukai and Watanabe estimated α to be 22.8 and 22.6 with and without exchange correlation corrections in the local pseudopotential theory retaining only first-order terms. Later these authors estimated $\alpha = 16$ by including higher-order terms in the pseudowave function in order to incorporate the Bloch character more accurately. Holtham and Jena[18] performed many orthogonalized plane-wave calculations and found anisotropy of about 10−20% in the values of α along different principal symmetry directions. An average value of $\alpha = 7.37$ is suggested for the Al matrix. Nevald et al.[8] used $\alpha = 27$ for the Al-based alloys. From the above results it appears that the calculated values of α give only an order of magnitude. In view of these uncertainties in α we assume in our calculations α as a parameter varying within the limits suggested by the above authors.

Equations (2), (4), (7), and (8) along with the tabulated values of various parameters in Table I are used to estimate q and η. We keep α between 7 and 20 which is the range as discussed above and make an attempt to obtain a suitable value of λ, such that the calculated values of q and η are in reasonably good agreement with the experimental values for the impurities of a particular row. The parameters (α, λ) are found to be (20,76), (7.37,−80), and (12,30) for the third-, fourth-, and fifth-row impurities, respectively, at the 1NN shell. The results for q and η at the 1NN shell along with experimental and other theoretical results are presented in Table

TABLE I. Physical parameters for dilute alloys of aluminum. Z_i is the chemical valency of the impurity, $a^{-1}da/dc$ is the fractional change in lattice parameter per unit concentration of the impurity, $\Delta\rho$ is the residual resistivity (in $\mu\Omega$ cm/at. %), Z_{eff} is the effective valence of the impurity, r_1 and r_2 (in a.u.) are the displaced positions of 1NN and 2NN, respectively, η_0 and η_1 are the phase shifts, $(\partial\eta_0/\partial k)_{k_F}$ and $(\partial\eta_1/\partial k)_{k_F}$ are the rate of change of phase shifts at the Fermi momentum k_F, and A, B, Φ, and ξ are the parameters that are determined from the phase shifts and their derivatives as described in the text.

Physical parameters	Impurities							
	Mg	Si	Cu	Zn	Ga	Ge	Ag	Cd
Z_i	2	4	1	2	3	4	1	2
$a^{-1}(da/dc)^a$	0.099	−0.042	−0.092	−0.013	0.045	0.045	0.023	−0.090
$\Delta\rho^a$	0.34	0.60	0.89	0.24	0.22	0.79	1.08	0.51
Z_{eff}	−1.602	1.255	−1.441	−0.921	0.274	0.726	−2.140	−0.453
r_1	5.449	5.362	5.332	5.380	5.416	5.416	5.402	5.333
r_2	7.650	7.607	7.592	7.616	7.634	7.634	7.627	7.592
η_0	−2.513	1.020	−1.994	−0.581	−0.477	1.122	−2.302	−0.781
η_1	−0.001	0.317	−0.090	−0.289	0.016	0.006	−0.353	0.023
$(\partial\eta_0/\partial k)_{k_F}$	−2.702	1.097	−2.144	−0.625	−0.513	1.206	−2.476	−0.840
$(\partial\eta_1/\partial k)_{k_F}$	−0.004	1.023	−0.289	−0.931	0.051	0.021	−1.139	0.076
A	0.030	0.031	0.052	0.019	0.025	0.045	0.075	0.038
B	0.068	0.180	0.099	0.151	0.020	0.033	0.238	0.025
Φ	3.767	−0.776	4.040	3.300	−0.432	4.282	3.274	−0.715
ξ	2.837	1.870	4.214	4.370	1.012	0.759	3.990	0.574

[a]Y. Fukai, Phys. Rev. 186, 697 (1969) and references therein.

II.

For the third-row impurities the calculated and experimental values of q and η are in good agreement for AlSi. However, the magnitude of the calculated q for AlMg is larger than the experimental value. For the fourth-row impurities Cu, Zn, Ga, and Ge in Al, the calculated q and η also show overall good agreement with the experimental values. The maximum deviation in q and η is for AlZn. For the fifth-row impurities Ag and Cd the agreement between the experimental and theoretical values of q and η is reasonably good except for η for AlCd. It is evident from Table II that the present results are better than the previous calculations.[7,8]

A considerable amount of work has been done for AlMg and AlZn alloys.[2,7−9,13,19] Therefore, we found it important to make further calculations for these alloys. We found that $(\alpha,\lambda)=(7.37,23)$ and $(7.37,−203)$ for AlMg and AlZn alloys, respectively, give close agreement with the experimental values for q and η (see Refs. c and d in Table II). Certainly these values of q and η are close to the respective experimental values for AlMg than those obtained by Prasad Rao and Mohapatra.[9] Interestingly, the value of α is the same as given by Holtham and Jena.[18] Assuming α evaluated by Holtham and Jena to be most reliable, we fixed $\alpha=7.37$ and varied λ to get an agreement between the calculated

and the experimental values of q and η for other alloys. q could be matched but observed values of η for all the alloys could not be explained.

Now we intercompare the results for the third-, fourth-, and fifth-row impurities in Al. It is evident from Table II that the Z-axis in Al alloys is in the perpendicular direction

$$|q_{ZZ}| \geq |q_{YY}| \geq |q_{XX}|$$

except for AlAg in which it is in the parallel direction. This is because of the fact that the valence- and the size-effect contributions have the same sign and therefore get added up in the parallel and perpendicular directions, while in the x direction these cancel each other. It is not possible to distinguish between the perpendicular and parallel directions in the NMR experiments.[7,20,21] However, neither the experiments nor our calculations predict the EFG axis along x direction.

The size effect dominates over the valence effect, except in an AlAg alloy where the valence-effect contribution dominates. In AlSi, AlGa, AlGe, and AlCd the asymmetry parameter η is small, therefore exhibiting nearly cylindrical symmetry in q. But in these alloys the size-effect contribution, which alone gives $\eta=0.33$, is large. On the other hand, AlAg has the highest value of η (equal to 0.31) among all the Al-based alloys and the valence-effect contribu-

TABLE II. q (in units of 10^{24} cm^{-3}) and η at 1NN shell for dilute alloys of aluminum with third-, fourth-, and fifth-row impurities. Parameters (α, λ) for the third-row impurities Mg and Si are (20,76), for the fourth-row impurities Cu, Zn, Ga, and Ge are (7.37,−80) and for the fifth-row impurities Ag and Cd are (12,30). Significance of q_{xx}, $q_\perp$, and $q_{||}$ is given in the text. Underlined values are the largest components of EFG's which define q_{zz} given in Eq. (9).

Impurities	EFG contributions	q_{xx}	$q_\perp$	$q_{||}$	q	η	Other theoretical results (q) Ref. 7[a]	Ref. 8[b]	Experimental values q	η
Mg	Total	0.288	−0.518	0.230	−0.518	0.11	−0.050	0.026	0.195	0.07
	Size	0.322	−0.484	0.161	(−0.159[c])	(0.07[c])				
	Valence	−0.034	−0.034	0.069						
Si	Total	−0.116	0.243	−0.127	0.243	0.05	0.119	0.058	0.282	0.03
	Size	−0.144	0.215	−0.072						
	Valence	0.028	0.028	−0.056						
Cu	Total	0.345	−0.497	0.152	−0.497	0.39	0.019	2.896	0.298	0.23
	Size	0.337	−0.505	0.168						
	Valence	0.008	−0.008	−0.017						
Zn	Total	0.042	−0.074	0.032	−0.074	0.14	−0.023	0.162	0.180	0.27
	Size	0.046	−0.069	0.023	(−0.181[d])	(0.25[d])				
	Valence	−0.004	−0.004	0.009						
Ga	Total	−0.127	0.266	−0.139	0.266	0.04	0.096	0.215	0.228	0.03
	Size	−0.157	0.236	−0.079						
	Valence	0.030	0.030	−0.060						
Ge	Total	−0.126	0.267	−0.141	0.267	0.06	0.148	0.324	0.328	0.03
	Size	−0.157	0.236	−0.079						
	Valence	0.031	0.031	−0.063						
Ag	Total	0.064	−0.140	0.204	0.204	0.37	0.042	−0.805	0.218	0.31
	Size	0.030	−0.046	0.015						
	Valence	−0.094	−0.094	0.188						
Cd	Total	0.029	0.279	−0.250	0.279	0.79		0.367	0.182	0.03
	Size	−0.123	0.185	−0.062						
	Valence	0.094	0.094	−0.187						

[a]Values of q including exchange correction.
[b]Values of q with Blatt correction.
[c]Calculated values of q and η with (α, λ) having values (7.37, 23).
[d]Calculated values of q and η with (α, λ) having values (7.37, −203).

tion (which exhibits cylindrical symmetry) dominates the size-effect contribution. Therefore, the nearly cylindrically symmetric behavior of the EFG tensor in some of the Al alloys is not necessarily because of the large valence effect as expected at the first instance and vice versa. The overall behavior of the Al matrix is such that the strain produced by the impurity, i.e, a^{-2}, $a^{-1}(da/dc)$, is small. Therefore, higher values of λ, at the 1NN, are needed for the proper size effect. This is because of larger screening of impurities in the high-electronic density matrix Al. At the 2NN shells the values of (α, λ) for the third-, fourth-, and fifth-row impurities are (7.37,11), (7.37,−5), and (7.37,5), respectively. The calculated and experimental values of q are given in Table III. Because of cylindrical symmetry at 2NN, $\eta = 0$ for all the alloys. Other theoretical results are also given there for comparison. In AlMg, AlSi, AlCu, AlZn, and AlAg, the size- and valence-

effect contributions are of opposite sign, therefore they cancel each other, while in AlGa, AlGe, and AlCd these are added up. The definite experimental value of EFG exists only for AlGe and AlAg alloys and our calculated results are in good agreement with them. For other alloys only an upper limit of the experimental EFG is known and therefore a quantitative comparison is not possible. However, the calculated values of q are of the correct order of magnitude and show the same trend as found in the experimental values. Comparing with the results of Fukai and Watanabe[7] and Nevald et al.,[8] our results are in closer agreement with the experimental values. This indicates that the size-effect contribution is important in quantitative explanation of experimental EFG.

By definition the EFG's are the second derivatives of the potential at nth NN and this potential arises due to change in electronic charge distribu-

TABLE III. q (in 10^{24} cm^{-3}) at 2NN in dilute alloys of aluminum. The parameters (α, λ) for third-, fourth-, and fifth-row impurities are (7.37,11), (7.37,−5), and (7.37,5), respectively. $q_{||}^S$ and $q_{||}^V$ have the same meanings as in text.

Impurity	$q_{||}^S$	$q_{||}^V$	q	Other theoretical results for q Ref. 7[a]	Ref. 8[b]	Experimental q
Mg	−0.034	0.021	0.013	0.029	−0.060	<0.020
Si	0.015	−0.005	0.010	−0.021	0.131	<0.020
Cu	−0.015	0.056	0.041	0.063	−0.160	<0.020
Zn	−0.002	0.025	0.023	0.023	−0.050	<0.020
Ga	0.007	0.002	0.009	−0.002	−0.018	<0.020
Ge	0.007	0.040	0.047	−0.021	0.055	0.031
Ag	−0.004	0.050	0.046	0.071	0.115	0.033
Cd	0.014	0.017	0.031		−0.028	

[a]Values of q including exchange corrections.
[b]Values of q with Blatt correction.

tion and elastic interactions in the matrix because of the inserted impurity. In general, we calculate $q_{||}$ the component of the EFG tensor along the line joining the host and impurity and estimate q_{xx} and $q_\perp$ assuming the rotational symmetry along the Z axis of EFG coordinates. In the AlMg alloy the Z_{eff} is negative and dilation is positive. The components of the EFG tensor q_{ij} arising due to both the electrostatic and elastic interactions are positive and therefore $q_{||}$ is also positive. This includes that at 1NN both the electrostatic and elastic interactions are repulsive along Z direction. According to symmetry considerations these interactions are attractive in the perpendicular direction and are of the opposite sign in the x direction. Therefore, the electrostatic and elastic fields around the impurity are anisotropic. At 2NN the nature of electrostatic interaction is repulsive while the elastic interaction is attractive and the resultant interaction remains repulsive. This concludes that the nature of elastic and electrostatic interactions is also at variance as one moves away from the impurity. Also the signs of q^V and q^S are not the same as that of Z_{eff} and $a^{-1}da/dc$.

In AlCu and AlZn, $a^{-1}da/dc$ and Z_{eff} are negative while in AlAg these are of opposite signs. For all these alloys except AlCu the nature of valence- and size-effect contributions components at 1NN is the same as found in AlMg in the parallel and perpendicular directions. At the 2NN, the valence-EFG tensors are positive while the size-EFG tensors are negative for all these alloys. This systematic is not consistent with that found at 1NN.

In an AlSi alloy the impurity has the positive charge while dilation is negative. The components of an EFG tensor along the parallel direction are

negative for both electrostatic and elastic interactions. These interactions are repulsive along the perpendicular direction while these are of opposite nature along the x direction. It denotes that the anisotropy of elastic and electrostatic fields for AlSi is of an opposite nature as that found for AlMg alloy. The lattice dilation and impurity-effective charges are positive in AlGa and AlGe alloys while these are negative in an AlCd alloy. However, the nature of components of EFG tensors is the same as found in an AlSi alloy. This again shows that the nature of components of EFG does not depend on the sign of dilation and residue valence. At 2NN, $q_{||}^S$ and $q_{||}^V$ are of the same sign as in AlGa, AlGe, and AlCd alloys, while these are of an opposite sign in an AlSi alloy. Conclusively, it is noted that Z_{eff} and $a^{-1}da/dc$ are not the sole parameters to decide the sign of components of q, although a correlation is found between $|q|$ and $|Z_{eff}|$ if one also accounts for the bound state. The other parameters relating to phase shifts which account for scattering are quite significant in the calculations of $q_{||}$ and its components.

In paper I we correlated $|q|$ and η at the 1NN and $|q|$ at the 2NN with the impurity-effective charge $|\Delta Z|$ in the Prakash and Lucasson[22] (PL) model. A systematic trend was observed. Sagalyn and Alexander have correlated $|q|$ and η with the nominal-valence difference. We made such an attempt to correlate $|q|$ and η with the effective charges on impurities calculated in the nominal-valence difference model and Z_{eff} tabulated in Table I and in the PL model. No correlation was found with nominal-valence difference and Z_{eff}. However, a correlation similar to that in paper I is found with $|\Delta Z|$ in the PL model.

In our calculations we evaluated the phase shifts satisfying the equations for residual resistivity and the Friedel sum rule. The expression for $\Delta\rho$ is based on a spherically symmetric scattering potential where the scattering is calculated at the spherical Fermi surface. However, the Fermi surface of an Al matrix is extended up to the third Brillouin zone and all the holes and pockets of the Fermi surface that may be important for $\Delta\rho$ are not accounted for. Holtham and Jena[18] have pointed out the anisotropy of the Bloch charge in the vicinity of an impurity; such anisotropy is also not accounted for in the present calculations. Again the preasymptotic part in the expression for $\Delta n(\vec{r})$ is an approximate one.

IV. CONCLUSIONS

In this paper we have adopted the Alfred and Van Ostenburg expression for the impurity-induced charge perturbation (which includes the preasymptotic contribution, although in an approximate manner) to evaluate the valence-effect contribution. The size-effect contribution is evaluated in the elastic continuum model. These two contributions are added up according to the required lattice geometry.

The core-enhancement factor α and the size-strength parameter λ are treated as fitting parameters for impurities of different rows in the Periodic Table. Systematically, the EFG and the asymmetry parameter η are explained. For multivalent matrix Al, it is expected at the first instance that the valence-effect contribution should dominate over the size-effect contribution. However, the calculations reveal that the size-effect contribution is larger than the valence-effect contribution for most of the alloys with non-transition-metal impurities. This conclusion is similar to that obtained in paper I for dilute alloys of Cu. The calculations of Ponnambalam and Jena[14] and Prasad Rao and Mohapatra[9] for Al vacancy and AlMg systems also found the dominating contribution to be due to size effect. However, a self-consistent calculation for both the valence- and size-effect contributions, especially within the preasymptotic region, is still required.

ACKNOWLEDGMENT

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