

## Strain field due to self-interstitial impurity in Ni

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**Abstract.** The embedded-atom method have been applied to study the strain field produced by the self-interstitial impurity at the octahedral site in Ni. The calculation have been carried out consistently on the basis of discrete lattice theory, using Kanzaki method. The atomic force constants are evaluated using Wills and Harrison interatomic potential. The dynamical matrix and external force are evaluated considering the interaction up to first nearest neighbors. The atomic displacements are tabulated up to 20NN's. These displacements are of oscillatory nature and of decreasing magnitude with NN's distance. The physical properties such as self-interstitial formation energy and volume change calculated using atomic displacements are in accordance with the earlier studies.

**Keywords.** Strain field; transition metal dilute alloys; point defects; Kanzaki method.

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### 1. Introduction

We had investigated the strain field in bcc (V, Fe, and Cr) [1,2] and fcc (Ni and Pd) [3] metals due to substitutional impurities using Kanzaki lattice static method based on discrete lattice theory. This method enables us to calculate the actual displacements from normal coordinates which are essentially the Fourier inverses of the direct space displacements. The data obtained from these calculations are of vital importance to study the elastic and electronic properties of dilute alloys [4–7].

In this paper the Kanzaki method has been extended to study the interstitial defects. The embedded atom method (EAM) has been applied to study the short range interactions due to self-interstitial Ni atom in Ni crystal. The atomic force constants are evaluated using Wills and Harrison potential. The paper is organized as follows: The necessary formalism and details of the embedded atom method is given in §2. The calculations and results are presented in §3 and these are discussed in §4.

### 2. Formalism

For a perfect crystal with self consistent pair potential  $\phi(r)$ , the total interaction energy  $\Phi_0$  is given as

$$\Phi_0 = \sum_n \phi(\vec{R}_n^0) \quad (1)$$

where  $\vec{R}_n^0$  is the equilibrium position of the  $n$ th host atom. If an impurity is introduced at the origin, the lattice gets strained, and the host atoms move to new equilibrium positions  $\vec{R}_n = \vec{R}_n^0 + \vec{u}(\vec{R}_n^0)$ , where  $\vec{u}(\vec{R}_n^0)$  are the atomic displacements. Kanzaki assumed that these displacements  $\vec{u}(\vec{R}_n^0)$  are produced by an appropriate distribution of external forces in the crystal which depend upon the nature of the impurity. The potential energy of the strained lattice under applied external forces is expanded in powers series of the displacements which in the harmonic approximation is given as

$$\Phi = \Phi_0 - \sum_{n,\alpha} u_\alpha(\vec{R}_n^0) F_\alpha(\vec{R}_n^0) + \frac{1}{2} \sum_{n,\alpha} \sum_{n',\beta} u_\alpha(\vec{R}_n^0) u_\beta(\vec{R}_{n'}^0) \phi_{\alpha\beta}(n, n') \quad (2)$$

where  $\Phi_0$  is the potential energy of the perfect lattice, and the force components

$$F_\alpha(\vec{R}_n^0) = - \left. \frac{\partial \Phi}{\partial u_\alpha(\vec{R}_n^0)} \right|_{u_\alpha(\vec{R}_n^0)=0} \quad (3)$$

and the force constants

$$\phi_{\alpha\beta}(n, n') = \left. \frac{\partial^2 \Phi}{\partial u_\alpha(\vec{R}_n^0) \partial u_\beta(\vec{R}_{n'}^0)} \right|_{u_\alpha(\vec{R}_n^0)=u_\beta(\vec{R}_{n'}^0)=0} \quad (4)$$

Here  $\alpha, \beta$  ( $x, y, z$ ) denote Cartesian components,  $F_\alpha(\vec{R}_n^0)$  is the  $\alpha$  component of the external force applied on the atom  $R_n^0$  and  $\phi_{\alpha\beta}(n, n')$  are the force constants which obey the crystal symmetries. The equilibrium values of the  $u(R_n^0)$  are obtained by minimizing  $\Phi$  with respect to  $u_\alpha(R_n^0)$ . One finds

$$F_\alpha(\vec{R}_n^0) = \sum_{n',\beta} \phi_{\alpha\beta}(n, n') u_\beta(\vec{R}_{n'}^0). \quad (5)$$

Evidently the displacements can be evaluated if  $F_\alpha(\vec{R}_n^0)$  and  $\phi_{\alpha\beta}(n, n')$  are known.

In the Kanzaki lattice static method the displacements are expanded in normal coordinates

$$u_\alpha(\vec{R}_n^0) = \sum_{\vec{q}} Q_\alpha(\vec{q}) \exp(i\vec{q} \cdot \vec{R}_n^0), \quad (6)$$

where  $\vec{q}$  is a wave vector and the expansion coefficients  $\vec{Q}(\vec{q})$  are normal coordinates known in dynamical theory. Since we are considering a periodic superlattice of defects, the wave vectors  $q$  must satisfy periodic boundary conditions, and all such physically distinct  $q$  vectors will be contained within the first Brillouin zone.  $\vec{Q}(\vec{q})$  are in general, complex and, to ensure the reality condition for displacements, we must have

$$Q(-q) = Q^*(q), \quad (7)$$

where the asterisk stands for the complex conjugate. Using eq. (6), one gets the Fourier transform of the total energy  $\Phi$  of the strained lattice as

$$\Phi = \Phi_0 - \sum_{\alpha q} F_{\alpha}(\vec{q}) Q_{\alpha}(\vec{q}) + \frac{N}{2} \sum_{\alpha\beta} \sum_q \phi_{\alpha\beta}(\vec{q}) Q_{\alpha}(\vec{q}) Q_{\beta}(\vec{q}) \quad (8)$$

where

$$F_{\alpha}(\vec{q}) = \sum_n F_{\alpha}(\vec{R}_n^0) \exp(i\vec{q} \cdot \vec{R}_n^0), \quad (9)$$

and

$$\phi_{\alpha\beta}(\vec{q}) = \sum_{n-n'} \phi_{\alpha\beta}(n-n') \exp[-i\vec{q} \cdot (\vec{R}_n^0 - \vec{R}_{n'}^0)], \quad (10)$$

$N$  is the number of lattice points in the crystal.  $F_{\alpha}(\vec{q})$  and  $\phi_{\alpha\beta}(\vec{q})$  are the Fourier transforms of  $F_{\alpha}(\vec{R}_n^0)$  and  $\phi_{\alpha\beta}(n-n')$ , respectively. The equilibrium condition in Fourier space in conjunction with eq. (8) gives

$$\sum_{\beta} \left[ N\phi_{\alpha\beta}(-\vec{q}) Q_{\beta}(\vec{q}) - F_{\beta}(\vec{q}) \delta_{\alpha\beta} \delta_{-\vec{q},\vec{q}} \right] = 0. \quad (11)$$

Equation (11) gives three simultaneous equations for three components  $Q_{\beta}(q)$  for each value of  $q$ . If  $\phi_{\alpha\beta}(q)$  and  $F_{\beta}(q)$  are known, eq. (11) can be solved for  $Q(q)$  which, in turn, gives  $u_{\alpha}(\vec{R}_n^0)$  from eq. (5).

For a central ion-ion potential, the dynamical matrix is written as

$$\phi_{\alpha\beta}(n) = \left. \frac{\partial^2 \phi}{\partial r_{\alpha} \partial r_{\beta}} \right|_{r=R_n^0} = \frac{R_{n\alpha}^0 R_{n\beta}^0}{|\vec{R}_n^0|^2} (A_n - B_n + \delta_{\alpha\beta} B_n), \quad (12)$$

where

$$A_n = \left. \frac{\partial^2 \phi}{\partial r^2} \right|_{r=R_n^0}, \quad B_n = \left. \frac{1}{|\vec{R}_n^0|} \frac{\partial \phi}{\partial r} \right|_{r=R_n^0}. \quad (13)$$

In metallic crystals the ions are screened by the conduction electrons thereby decreasing the ionic potential faster, which exhibit oscillatory behavior at large distances. It has been found that in d-band metals the screening is very heavy [8–10]. Therefore, major contribution to  $\phi_{\alpha\beta}(\vec{q})$  and  $F_{\alpha}(\vec{q})$  in these metals is expected to arise from the few NN's. Including the interactions up to 1NN's,  $\phi_{\alpha\beta}(\vec{q})$  for fcc structure, from eqs (10) and (11) becomes

$$\phi_{\alpha\alpha}(\vec{q}) = 2(A_1 + B_1) \left[ 1 - \cos\left(\frac{q\alpha a}{2}\right) \left[ \cos\left(\frac{q\beta a}{2}\right) + \cos\left(\frac{q\gamma a}{2}\right) \right] \right] \quad (14)$$

$$\phi_{\alpha\beta}(\vec{q}) = 2(A_1 - B_1) \left[ \sin\left(\frac{q\alpha a}{2}\right) \sin\left(\frac{q\beta a}{2}\right) \right], \quad (15)$$

where  $\alpha \neq \beta \neq \gamma$  and  $a$  is the lattice parameter.

Since the interstitial impurity interacts with short-range interactions, the external force is expected to arise appreciably only from 1NN's contribution. The force obtained using eq. (8) evaluated at the 1NN shell of the interstitial impurity is

$$F_{\alpha}(q) = i2F_1 \sin(q_{\alpha}a), \quad (16)$$

where  $F_1$  is the force acting on the 1NN site of the interstitial impurity. With the knowledge of  $\phi_{\alpha\beta}(\vec{q})$  and  $F_{\alpha}(\vec{q})$ , eq. (11) is solved for  $\vec{Q}(\vec{q})$  for radial forces on the 1NN's of the impurity. These values of  $\vec{Q}(\vec{q})$  are used in eq. (6), summation is replaced by integration and  $\vec{u}(\vec{R}_n^0)$  are calculated for different values of  $\vec{R}_n^0$  (using quadrature method of numerical integration).

### 2.1 Estimation $F_1$ using embedded atomic potential

The interstitial impurities in fcc metal occupy octahedral site and their interaction with the host atom could be best described by short range potential or first principle calculations. One such method to describe the interactions in the transition metals is EAM. The short range interaction in TM can be described using EAM method. The EAM is based on the density-functional theory. In this method, energy required to place a small impurity atom in a lattice is taken solely as a function of electron density at that particular site. The energy of each atom is computed from the energy needed to embed the atom in the local-electron density as provided by the other atoms in the metal. This electron density is approximated by the superposition of atomic-electron densities. The total energy of the system is

$$\Phi_{\text{total}} = \sum_i F_i(\rho_{hi}) + \frac{1}{2} \sum_{i=j, i \neq j} \phi_{ij}(R_{ij}). \quad (17)$$

$F_i(\rho_{hi})$  is the energy to embed atom  $i$  in an electron density  $\rho_{hi}$  and  $\phi_{ij}$  is the two body central potential between  $i$  and  $j$ .  $\rho_{hi}$  is the electron density at the atom  $i$  due to all other atoms.

$$\rho_{hi} = \sum_{j(\neq i)} f_j(R_{ij}) \quad (18)$$

$f_j$  is the electron density of atom  $j$  as a function of distance from its center.  $R_{ij}$  is the distance between atom  $i$  and  $j$ . The embedding function is universal, it does not depend on the source of the background density. Thus same embedding function is used to calculate the energy of an alloy that is used in the pure metal.

To use embedded atom method in a lattice model calculation, the  $f$  and  $F$  functions must be specified for such atomic species. In the EAM calculations, the embedding function is determined by complex fitting procedure. Foiles [11] has recently introduced a straightforward scheme which is also used in the present calculations. The pair-interaction term  $\phi_{ij}(R_{ij})$  is purely repulsive and is considered of the form

$$\phi_{ij}(R_{ij}) = \frac{Z_i(R_{ij})Z_j(R_{ij})}{R_{ij}} \quad (19)$$

where  $Z(R)$  is parameterized to

$$Z(R) = Z_0(1 + \beta R^v) \exp(-\alpha R). \quad (20)$$

The value of  $Z_0$  is the number of outer electrons of the atom and for Ni,  $Z_0 = 10$ . The other parameters,  $\alpha, \beta$  and  $v$  are determined empirically. It was found that  $v = 1$  leads to a good representation of elastic constants for Ni. Rest of the parameters have been determined for Ni, so as to yield the elastic constants and vacancy-formation energy of each material as well as the dilute limits of the heat of solution of the binary alloys [11]. The external force calculated from eq. (17) is of the form

$$F_\alpha(R_n^0) = - \left. \frac{\partial F}{\partial \rho} \frac{\partial \rho}{\partial R} \right|_{R=R_n^0} - \frac{1}{2} \left. \frac{\partial \phi}{\partial R} \right|_{R=R_n^0}. \quad (21)$$

The slope of the embedding function and density are evaluated from the variation of embedding function with density and variation of density with distance, which are explicitly solved for Ni.

### 3. Calculations and results

The Kanzaki method was applied to calculate strain field due to self-interstitial Ni atom in Ni crystal. Some of the physical parameters, force constants  $A_1$  and  $B_1$  calculated from the Wills and Harrison potential [3] and the calculated external force using embedded atom method (EAM) are given in table 1. The physical parameters for defining the pair-interaction in EAM are given in table 2.

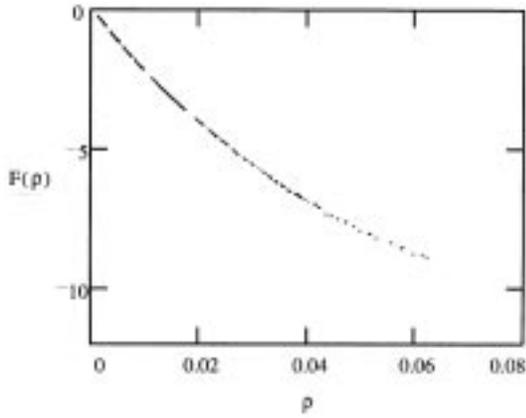
The embedding function as a function of density for Ni are derived empirically from the data in [11] which is shown in figure 1. The atomic density of Ni are calculated from the spherically averaged s and d like densities which are computed from Clementi and Roetti [12]. The variation of the atomic density with distance are plotted in figure 2. The total energy is defined using eq. (17) and external force using eq. (21). The interstitial impurity experiences short range interactions with host atoms. Therefore, it is adequate to assume that the defect exerts an appreciable force only on its first nearest neighbors and so force  $F_1$  can be evaluated at the 1NN's.

**Table 1.** Some physical parameters (all in a.u.) for Ni.  $a$  is the lattice constant,  $\Omega_0$  is the equilibrium volume,  $A_1$  and  $B_1$  are the force constants and  $F_1$  is the external force.

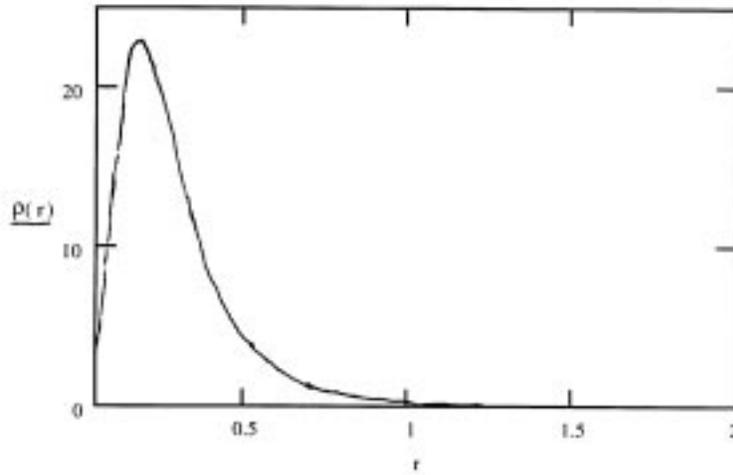
Host	$a$	$\Omega_0$	$A_1$	$B_1$	$F_1$
Ni	6.65	90	0.028	-0.00089	0.252

**Table 2.** Physical parameters used to define the effective charge for the pair interactions between Ni-Ni atoms.  $Z_0$  is the outer electrons,  $\alpha, \beta$ , and  $v$  are empirical parameters.  $n_s$  is the number of s electrons in the outer orbit of Ni.

$Z_0$	$\alpha$	$\beta$	$v$	$n_s$
10	1.2950	0.0595	1	0.8478



**Figure 1.** The variation of embedding function  $F(\rho)$  with background density for Ni. The embedding energy is in eV, electron densities in  $\text{\AA}^{-3}$ .



**Figure 2.** The variation of atomic densities  $\rho$  vs.  $r$  for Ni, computed using densities of s and d electrons, which are associated with their Hartree–Fock wave functions. The atomic densities are in  $\text{\AA}^{-3}$  units and  $r$  is in  $\text{\AA}$ .

These values of  $F_1$  and the calculated values of  $A_1$  and  $B_1$ , are used to calculate  $\phi_{\alpha\beta}(\vec{q})$  and hence  $\vec{Q}(\vec{q})$  with the help of eq. (11). The inverse Fourier transform of  $\vec{Q}(\vec{q})$ , as given in eq. (6), gives  $\vec{u}(\vec{R}_n^0)$ . The numerical calculations are simplified if we integrate over the cube of edge  $4\pi/a$  which inscribes the first Brillouin zone (BZ) and using the fact that, for any function  $F(q)$

$$\int_{\text{BZ}} F(q) dq = \frac{1}{2} \int_{\text{cube}} F(q) dq \quad (22)$$

for fcc structures. The integration is carried out by the Gaussian quadrature method.

**Table 3.** Atomic displacements of the NN's of Ni self-interstitial impurity. The displacements are expressed in terms of lattice constant  $a$ .  $(n_1, n_2, n_3)$  are the coordinates of NN's and  $(u_x, u_y, u_z)$  are the Cartesian coordinates of the displacements.

NN's $(n_1, n_2, n_3)$	Displacement components			
	$u_x$	$u_y$	$u_z$	$ u $
100	0.2465	0.0000	0.0000	0.2465
111	0.0047	0.0047	0.0047	0.0081
210	0.0392	0.0325	0.0000	0.0509
221	0.0103	0.0103	0.0090	0.0172
300	0.0054	0.0000	0.0000	0.0054
311	0.0136	0.0078	0.0078	0.0175
320	0.0135	0.0135	0.0000	0.0191
331	0.0073	0.0073	0.0041	0.0111
333	0.0052	0.0052	0.0052	0.0090
410	0.0034	0.0025	0.0000	0.0043
421	0.0067	0.0054	0.0029	0.0090
430	0.0065	0.0065	0.0000	0.0092
432	0.0053	0.0047	0.0036	0.0079
441	0.0046	0.0046	0.0017	0.0068
443	0.0037	0.0037	0.0031	0.0061
520	0.0027	0.0023	0.0000	0.0035
522	0.0039	0.0024	0.0024	0.0052
533	0.0037	0.0026	0.0026	0.0052
540	0.0037	0.0035	0.0000	0.0051
544	0.0028	0.0022	0.0022	0.0042
630	0.0023	0.0015	0.0000	0.0027
641	0.0027	0.0018	0.0005	0.0033

The atomic displacements due to self-interstitial impurity, in units of lattice constant, are tabulated in table 3. The 1NN shows maximum displacement of 0.24, away from the self-interstitial Ni atom. The other NN's are also displaced away from the impurity atom, i.e., the lattice shows expansion. The magnitude of these displacements decreases oscillatory with NN distance. The calculated value of the self-interstitial formation volume, i.e.,  $\Delta V/\Omega$  is 1.87 whereas its value obtained from earlier work is 2.11 [11]. The self-interstitial formation energy or relaxation energy is given by

$$E_r = -\frac{1}{2} \sum_{n,\alpha} u_{n\alpha} F_{n\alpha}, \quad (23)$$

where  $F_\alpha$  is isotropic in nature and the atomic displacements of 1NN's from table 3 are used to evaluate relaxation energy. The relaxation energy comes out to be 5.58 eV whereas earlier calculated value is 5.05 eV [11]. Since the values of relaxation energy and change in volume are in agreement with the values from earlier work, it can be concluded that our calculation using discrete lattice theory is consistent with the actual description of the atomic displacements of the defected system.

#### 4. Discussion

We have used here embedded atom method [11] for the self-interstitial impurity in Ni. The embedded atom is based on background density of the host and has been applied to

the transition metal, to describe various properties such as bulk modulus, lattice constants, force constants etc. The universality of the embedding function makes it a valuable tool to study the transition metal based alloys. The lattice displacements due to interstitial impurities are difficult to evaluate due to lack of appropriate potential for the system as most of the potential fails in this range. The reasonable agreement in the calculated results with the earlier work justifies that results obtained using the discrete lattice model for interstitial defects are consistent with the potential considered and the more exact description of the interatomic interactions would certainly improve the results.

In the numerical calculations the cubic symmetry of the lattice is retained although the exact anisotropy of the Brillouin zone is not accounted for. This may not introduce serious error considering other simplifications in the calculations.

The tabulated values of displacements may be quite useful to investigate heat of solution, electric field gradients, asymmetry parameter, wipe out number, Knight shift and other properties of the defect lattice where impurity induced displaced positions of the host atoms in dilute alloys of Ni are needed. This will help in the basic understanding of the alloy formation. Further, these study will explain the strength at high temperature, high stiffness, low coefficient of thermal expansion and chemical compatibility in a variety of environments.

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