

DYNAMICAL PROPERTIES OF POINT DEFECTS IN METALS

ABSTRACT

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SUMMARY

DYNAMICAL PROPERTIES OF POINT DEFECTS IN METALS

The study of dynamics of irradiation-produced point defects: self-interstitial atoms (SIAs) and vacancies is of fundamental importance in understanding of various physical properties of irradiated metals. Of special interest, is the vibrational frequency spectra of the defect and a few of its neighbours. The presence of point defects in crystal is reflected on the vibrational frequency spectra, which are different from those of the ideal lattice atoms. The present thesis discusses the dynamical properties of self interstitials and vacancies in cubic metals. A semi-empirical potential, called the embedded atom method (EAM) potential in fcc metals has been determined. The local frequency spectra of SIAs in Cu, which is a typical fcc metal, is discussed on the basis of this new potential. The local frequency spectra of SIAs and vacancies in bcc metals are also discussed on the basis of the Johnson and Wilson (JW) potentials [1].

Chapter I contains a brief introduction to the thesis emphasising the generation of the EAM potentials in fcc metals and also various physical properties of the solid containing point defects. The advent of new potentials like EAM potentials or N-body potentials replaces the empirical pair potentials and helps to calculate many properties of metals where the electronic contribution plays important role. Regarding the vibrational frequency spectra of SIAs, a number of characteristic defect modes: the low-frequency resonance modes and the high-frequency localized modes are discussed. The effect of resonance modes on some thermodynamic properties and elastic constants of the defect metals are pointed out. The local frequency spectra of the first and second neighbours of a vacancy do not show the presence of characteristic modes but the frequency spectra of the first neighbours shift towards the lower frequency region of the spectrum. Formation entropy of a vacancy is also discussed.

In chapter II we have generated the EAM potentials for fcc metals. These potentials are based on the electron density functional theory developed by Daw and Baskes [2]. The EAM potential consists of two parts: (i) the embedding function, which is the energy required to embed an atom in the field of the other atoms and (ii) the pair part, from the repulsion of atomic cores, which is of the Morse form. The new potential can explain various types of perfect crystal properties as well as defect properties in metals like vacancies, crack-tips, alloys, surfaces etc. In the present work the embedding function has been chosen from the one used by Banerjea and Smith [3]

$$F(\rho) = F(\rho_e) \left[1 - \gamma \ln \left(\frac{\rho}{\rho_e} \right) \right] \left(\frac{\rho}{\rho_e} \right)^\gamma \quad (1)$$

where ρ is the electron density at a lattice site which is due to the contribution from all other atoms, except the particular atom at the lattice site. The pair part is

$$\Phi(r) = D \left[e^{-2\alpha(r-r_0)} - 2e^{-\alpha(r-r_0)} \right] \quad (2)$$

The different potential parameters γ , D , α , r_0 are fitted to elastic constants, vacancy formation energy, cohesive energy and lattice constant, in a third neighbour model. The generation of the EAM potentials becomes complete when all the potential parameters are obtained. The generated potentials are then used to discuss the divacancy binding energy, the low-indexed plane surface energies and phonons for six fcc metals : Cu, Ag, Au, Ni, Pd and Pt. The obtained phonons in the present study are in good agreement with experimental phonons in Cu, Ag and Ni but in the case of Au, Pd and Pt the agreement is not so good. The obtained phonons are then used to discuss the density of state and thermal displacement of an atom in Cu.

In chapter III we have introduced the density of states, the Greens function and the local density of states. The density of state $Z(\omega)$ is given by

$$Z(\omega) = \frac{1}{3N} \sum_{\mathbf{k}\sigma} \delta(\omega_{\mathbf{k}\sigma} - \omega) \quad (3)$$

where $\omega_{\mathbf{k}\sigma}$ are phonons frequencies and N is the number of atoms in

the solid. The density of state is also expressed in terms of Green's function as [4]

$$Z(\omega) = \frac{2M\omega}{3N\pi} \operatorname{Im} \sum_{\alpha} G_{\alpha\alpha}(1,1;\omega) \quad (4)$$

where $G_{\alpha\alpha}(1,1;\omega)$ is the diagonal element of the same site Green's functions of the lattice. The local density of state of an atom at site 1 in the α -direction is

$$Z_{\alpha}(1,\omega) = \frac{2M\omega}{\pi} \operatorname{Im} G_{\alpha\alpha}(1,1;\omega) \quad (5)$$

The total spectrum of the lattice is the sum of local frequency spectra of all atoms in the lattice. The concept of the local density of states is quite useful in the study of defect dynamics because the vibrational behaviour of the defect and its neighbours are affected most so that the local density of states of the defect site show the excitations of some characteristic defect modes : the low-frequency resonance modes and the high-frequency localized modes.

The local density of states are quite useful in calculating the various properties of the defect crystals. The change in various thermodynamic quantities of a crystal can be expressed in terms of the local density of states easily. The mean square thermal displacement is expressed as

$$\langle u_{\alpha}^2 \rangle = \int_0^{\alpha} \frac{Z_{\alpha}(1, \omega)}{M\omega^2} \varepsilon(\omega, T) d\omega \quad (6)$$

where $\varepsilon(\omega, T) = \frac{\hbar\omega}{2} \coth\left(\frac{\hbar\omega}{2kT}\right)$.

The vacancy formation entropy can be expressed in term of a change in the frequency spectra and is give by :

$$S_{1V}^F = k \int_0^{\alpha} \sigma(\omega, T) \Delta Z(\omega) d\omega = k \int \sum_d (Z(d, \omega) - Z(\omega)) \sigma(\omega, T) d\omega \quad (7)$$

where $\sigma(\omega, T) = 1 - \ln\left(\frac{\hbar\omega}{kT}\right)$, and d stands for neighbours of vacancy.

In Chapter IV the local density of states of $\langle 100 \rangle$ -dumbbell has been discussed in fcc metals, using Green's function method, with copper as a typical example. The EAM potential has been used to calculate the ideal lattice force constants and the force constants changes in the defect space, using a second neighbour force model. The defect space is divided into different irreducible representations and many characteristic modes have been obtained at different frequencies in THz: the resonance modes are E_g (2.74), A_{2u} (5.56) and E_u (3.12); the localized modes are A_{1g} (11.08), E_g (12.94), E_u (13.16). But when the experimental phonons based force constants are used in the calculations of the ideal lattice Green's functions, and the EAM potential is used to calculate the defect lattice force constants, the resonant frequencies are E_g (1.62), E_u (2.20) and the localized modes are A_{1g} (7.30, 14.06), A_{2u} (7.36). The use of EAM potential in the dynamic-defect calculations has been done for the first time and could reproduce the defect modes. The presence of low frequency resonance modes also explains the reduction in shear moduli and the long-range migration of the SIAs at low temperatures.

In Chapter V we have calculated the local density of states of $\langle 110 \rangle$ -dumbbell in bcc metals: α -Fe, Mo and W using the Green's function method. Two sets of force constants have been used to calculate the ideal lattice green's function (i) from the (JW) potentials and (ii) from the experiments based phonons. The force constant changes in the defect space have been calculated from the JW potentials. In both the cases many characteristics defect modes have been obtained at different frequencies when the JW potentials are used consistently the resonance modes occur at : B_{1g} (1.68), B_{1u} (2.18), B_{2u} (2.70), B_{3u} (0.59) and the localized modes at A_g (11.32), B_{1g} (11.32), B_{2u} (11.70), B_{3u} (16.88) for α -Fe; similarly the resonance modes B_{2g} (2.16), B_{1u} (3.28), B_{2u} (2.64), B_{3u} (2.16) and the localized modes at A_g (9.70, 16.28), B_{1g} (9.38, 15.62), B_{1u} (15.86), B_{2u} (9.58), B_{3u} (13.72) for Mo; also the resonance modes B_{2g} (1.14), B_{1u} (2.28), B_{2u} (1.82), B_{3u} (1.38) and the localized modes at A_g (8.04, 13.92), B_{1g} (8.14), B_{2g} (13.54), B_{1u} (13.72), B_{2u} (8.30), B_{3u} (11.76) for W. When the experimental phonons are used, the resonance modes are: for α -Fe, B_{1g} (2.42), B_{2g} (1.86), B_{1u} (3.08), B_{2u} (3.26), B_{3u} (1.88) and the localized

modes at A_g (9.37, 10.99), B_{1g} (10.94), B_{2u} (11.24), B_{3u} (9.46, 16.10);
 for Mo the resonance modes B_{1g} (1.39, 7.995), B_{2g} (2.73), B_{1u} (3.55),
 B_{2u} (3.11), B_{3u} (2.59) and the localized modes at A_g (8.26, 12.77),
 B_{2g} (12.17), B_{1u} (12.38), B_{2u} (8.07), B_{3u} (10.965); for W the
 resonance modes B_{2g} (1.98), B_{1u} (2.78), B_{2u} (2.18), B_{3u} (1.86) and
 the localized modes at A_g (7.30, 12.12), B_{1g} (7.34), B_{2g} (11.84),
 B_{1u} (12.00), B_{2u} (7.46), B_{3u} (10.40).

The obtained local frequency spectra are then utilized to
 calculate the mean square thermal displacements of the dumbbell,
 in all the three metals and with both sets of force constants
 used. The results show an increase in thermal displacements as
 compared to that of the host atom, especially in the case of
 α -Fe. This is the result of low-frequency resonance modes. The
 resonance modes also explain the long range migration of SIAs at
 low temperatures.

In Chapter VI we have calculated the local frequency spectra
 of the first and second neighbours of vacancies in α -Fe, Mo and W
 using the same Green's function's method and following the same

procedures regarding the force constants and force constant changes as done in the case of SIAs. It has been found that there is a significant shift of the density of states towards the lower frequency in the spectra of the first neighbours as found by Hatcher *et al* [5] in Cu, whereas the spectra of second neighbours resemble those of the respective host spectra in the case of Mo and W but in the case of α -Fe the second neighbours' spectra shift towards the higher frequencies when the lattices are allowed to undergo relaxation. The vacancy formation entropy also are calculated from the local frequency spectra in all the three metals for the case of relaxed lattices and also for unrelaxed lattices. The values of the vacancy formation entropy for relaxed lattice when the JW potentials are used consistently are : α -Fe (1.559k), Mo(2.25k), W(3.199k), but when the experimental based phonons are used, we get α -Fe(1.485k), Mo(1.94k) and W (2.446k). In the case of unrelaxed lattices the values are: α -Fe(2.043k), Mo(1.61k), W(1.921k) when JW potential is used and they are α -Fe(1.646k), Mo(1,69k), W(2.143k) when experimental phonons are used. In general, the obtained values of formation entropy are comparable to those obtained by others.

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