BUBBLES AND VOIDS IN THE ELECTRON MICROSCOPE

A thesis submitted for the degree of
Doctor of Philosophy,
of the University of Surrey, by
Brian Cochrane
Abstract

In this work a high voltage electron microscope has been used to produce and image voids in nickel. From these experiments the dislocation bias has been estimated to be 6.5% and it has been shown that voids also have a bias of about 0.5% for interstitials over vacancies.

A computer program has been written which allows investigation of the strain field existing in the matrix material surrounding voids and bubbles. This program simulates the electron microscope contrast of defects using the two-beam and column approximations and can incorporate realistic pore shapes and strain fields. Results are presented both making the assumption that the strain fields are spherically symmetric and not assuming this constraint. In the latter case a Green's function method has been applied to calculate the defect strain field. This program has been used to study the effects on the image of varying the parameters involved such as defect depth, size and shape, foil thickness and orientation, diffraction vector and deviation from the Bragg position. Images are displayed at the in focus and defocussed image planes and are compared with experimental micrographs of helium bubbles in vanadium and niobium/1% zirconium. The simulated images agree well with the experimental images and have shown that the extent of the strain contrast is very limited even at large g and with high gas pressure in the pore. Experimental images are often taken with the microscope defocussed in order to enhance the pore edges. It is demonstrated that the small strain contrast will be reduced and may even be lost completely when the microscope is operated in this condition.
Acknowledgements

The author would like to thank the Central Electricity Generating Board for the use of the facilities at Berkeley Nuclear Laboratory and in particular Dr. S. B. Fisher in connection with work carried out there. Provision of graphics packages at Queen Elizabeth College by A. F. Clark and N. R. Arnot has helped in the production of this thesis and useful discussions with Dr. S. K. Tyler, Dr. R. F. Scott and Dr. R. Bullough are acknowledged. Help and suggestions from Dr. P. J. Goodhew have been greatly appreciated as has the provision of facilities by the Micro-structural Studies Unit at Surrey University. Finally the author would like to thank Kerry for all the encouragement, help and support given during the course of this project.
<table>
<thead>
<tr>
<th>Symbol</th>
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<tr>
<td>$\beta'$</td>
<td>Fractional burn-up</td>
</tr>
<tr>
<td>$\sigma_f$</td>
<td>Fission cross section</td>
</tr>
<tr>
<td>$E_{\text{max}}$</td>
<td>Maximum energy transfer</td>
</tr>
<tr>
<td>$m_n$</td>
<td>Neutron mass</td>
</tr>
<tr>
<td>$M$</td>
<td>Mass of target atom</td>
</tr>
<tr>
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<td>Original neutron energy</td>
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<tr>
<td>$E_d$</td>
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<tr>
<td>$E_{\text{int}}(r, \theta)$</td>
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<td>$J$</td>
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</tr>
<tr>
<td>$D_\beta$</td>
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</tr>
<tr>
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<tr>
<td>$b$</td>
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<td>$\Delta V$</td>
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</tr>
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<tr>
<td>$E_{\text{v}}, E_{\text{i}}$</td>
<td>Migration energy of vacancy and interstitial respectively</td>
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<tr>
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<tr>
<td>$\xi'_j$</td>
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<td>$e$</td>
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<td>Symbol</td>
<td>Meaning</td>
</tr>
<tr>
<td>--------------</td>
<td>---------------------------------------------------</td>
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<td>( \phi_j (z) ) ( \phi_k (z) )</td>
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<tr>
<td>h,k,l</td>
<td>Orientation of foil</td>
</tr>
<tr>
<td>D_e</td>
<td>Defect depth</td>
</tr>
<tr>
<td>T(p)</td>
<td>Amplitude transfer function</td>
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<td>B(p)</td>
<td>Aperture function</td>
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<td>W(p)</td>
<td>Wave aberration function</td>
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<td>2-D vector in Fourier space</td>
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<td>Spherical aberration</td>
</tr>
<tr>
<td>C_a</td>
<td>Axial astigmatism</td>
</tr>
<tr>
<td>C_c</td>
<td>Chromatic aberration coefficient</td>
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<tr>
<td>( \Delta E )</td>
<td>Full width at half height of electron energy distribution</td>
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<tr>
<td>( \delta_{ij} )</td>
<td>Kronecker delta</td>
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<tr>
<td>L</td>
<td>Elasticity constant used in Green's function calculation</td>
</tr>
<tr>
<td>R_x</td>
<td>Loop interaction radius</td>
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CHAPTER ONE

IRRADIATION PRODUCED VOID SWELLING
Introduction

The advancement of nuclear power production technology from Magnox to gas cooled to breeder reactors has been accompanied by problems for the engineer.

Development of the breeder reactor is a result of the desire to make more efficient use of the world's uranium reserves. Gas cooled reactors which use enriched uranium as fuel have a conversion ratio of less than unity. This means that the number of fissile atoms produced from the fertile material is less than the number of fissile atoms of fuel destroyed in the process. Thus there is a net consumption of the fissionable uranium isotope $^{235}$U in the fuel cycle. This isotope occurs as less than 1% of the available uranium. Breeder reactors will allow use of about 75% of the uranium reserves.

The main worldwide emphasis has been on the development of the liquid sodium cooled fast breeder reactor (LMFBR). These reactors, for example at Dounreay, use a mixture of uranium and plutonium oxides for fuel with pellets of $^{238}$U$_2$O$_2$ above and below the mixed oxide fuel. The axial blanking regions improve overall breeding by capturing neutrons leaking from the core. In the core fissile $^{239}$Pu is produced from non-fissile $^{238}$U by neutron capture. Figure 1.1 shows a typical configuration of a LMFBR fuel system.

The material used for fuel cladding is 316 austenitic stainless steel with a maximum mid-wall cladding temperature of about 600°C. The main duty of the cladding alloy is that it must remain
UO$_2$ Insulator pellets

Pall 90% of theoretical density
diam. = 0.5 cm

90-cm Mixed oxide pellet stack

end cap bottom

Inconel reflector

316 SS Plenum spacer

Tag-gas capsule

end-cap top

pull through wire attachment

Wrap wire 316 SS

Cladding, 316 SS

pellet to cladding gap = 0.14 mm
length = 240 cm

Figure 1.1 Fuel pin of a fast test reactor (Olander (1))
leak tight throughout its dwell time in the reactor. This ensures that active fission products are not released into the primary cooling circuit. Thus the cladding material must conform to strict specifications. It must be compatible with both fuel and coolant at high temperatures; the mechanical properties must be such as to prevent failure during steady state and transient reactor operations and the cladding must also have sufficient strength and corrosion resistance to remain intact during fuel handling and storage operations.

One of the problems accompanying the advance of reactor technology is an increase in radiation damage occurring in the core region of the reactor, e.g. to the fuel cladding material. This was not a significant problem with thermal reactors but it is with fast reactors. Thermal reactors utilize neutrons with kinetic energy of a few electron volts, which are known as slow or thermal neutrons. At 300K a thermal neutron has a kinetic energy of approximately 0.025eV. Fast reactors use neutrons with kinetic energy greater than 0.1MeV known as fast neutrons. Table 1.1 is a comparison of typical 1000 MW thermal and fast reactor performance characteristics.

From the table it is clear that there are several important differences in the characteristics of thermal and fast reactors.

1. The flux in a fast reactor is a factor of one hundred times larger than that in a thermal reactor. This large increase in flux allows the fuel to produce more power per unit volume despite fission cross sections that are smaller in fast reactors.

2. The average burn up is about three times as large in a fast reactor as in a thermal reactor. The fractional burn up, $\beta'$, is given
<table>
<thead>
<tr>
<th></th>
<th>THERMAL REACTOR</th>
<th>FAST BREEDER</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fissile species enrichment</td>
<td>3% $^{235}\text{U}$ in $^{239}\text{U}$</td>
<td>15% $^{239}\text{Pu}$ in $^{239}\text{U}$</td>
</tr>
<tr>
<td>Fission cross section $\sigma_f$/barns</td>
<td>550</td>
<td>1.8</td>
</tr>
<tr>
<td>Core averaged neutron -2 -1 flux / n cm s</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Thermal</td>
<td>$3 \times 10^{13}$</td>
<td>$1 \times 10^{11}$</td>
</tr>
<tr>
<td>Fast($&gt;0.1\text{MeV}$)</td>
<td>$5 \times 10^{3}$</td>
<td>$8 \times 10^{15}$</td>
</tr>
<tr>
<td>Burn up / %</td>
<td>3</td>
<td>10</td>
</tr>
<tr>
<td>Fast fluence / n cm$^2$</td>
<td>$3 \times 10^{21}$</td>
<td>$3 \times 10^{23}$</td>
</tr>
<tr>
<td>Irradiation time(at full power) / years</td>
<td>2</td>
<td>1.5</td>
</tr>
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</table>

Table 1.1 Comparison of typical 1000MW THERMAL and FAST reactors. (after Olander [1])
Higher burn up is economically necessary in a fast reactor to keep fuel fabrication and reprocessing and out of reactor inventory to a minimum.

3. The fast neutron fluence in a fast reactor is approximately one hundred times greater than in a thermal reactor.

It is this large difference in fast neutron flux and therefore fluence which is responsible for the radiation damage to the cladding and wrapping being a much more severe problem in fast, than in thermal reactors. In fast reactors core structural components can undergo volume changes of the order of +10%. All regions in the core do not operate under the same neutron flux and temperature conditions. The different radiation exposure and temperature results in differential swelling of components, and therefore warping, leading to servicing problems. It has been estimated that a reduction in the swelling of fuel element cans by a factor of three, from 15% to 5%, would give a saving over the first fifty years of the American programme of up to $4,000 million. This economic incentive has resulted in much work being carried out on various alloys in an attempt to both understand the fundamental reasons for swelling and to produce alloys which reduce this effect as much as possible.
1.1. Previous work

In 1966 Cawthorne and Fulton [2], working at Dounreay using transmission electron microscopy, discovered that the swelling occurring in cladding material was due to the formation of small cavities within the alloy. These cavities did not have sufficient gas contained in them to be classed as bubbles and are known as voids. Since this first discovery extensive investigations of the phenomenon have been carried out in a variety of metals and alloys, all of which swell by varying amounts. Norris [3] gives a list of materials which had been or were being studied in 1972. Much work has been carried out since that date, but the range of materials is still largely encompassed by that list which includes: aluminium, copper, nickel, stainless steels, iron-nickel alloys, irons, vanadium, niobium, tantalum, tungsten, magnesium and cobalt.

Swelling occurs as a result of particle collisions. When a bombarding particle strikes a lattice atom kinetic energy is transferred. Neutrons, having no charge, do not exert any appreciable force on an atom unless they strike the nucleus. Usually the probability of a fast neutron colliding elastically is greater than that of such a neutron colliding inelastically. The maximum energy transferred, $E_{\text{max}}$, in an elastic collision is given by [4]:

$$E_{\text{max}} = \frac{4m_nM E_n}{(M + m_n)^2}$$

where $m_n$ is the neutron mass

$M$ is the mass of the target atom
$E_n$ is the original neutron energy.

If the energy transferred is greater than some threshold value, $E_d$, then the lattice atom will be displaced from its lattice site. This threshold value, called the displacement energy, is direction dependent, but of the order of 25eV to 30 eV in most metals. From the equation (1.1), it can be seen that for an atom of mass 10 a.m.u. a neutron would have to have an initial energy of about 75eV to displace the atom. As thermal neutrons have energies less than 0.1 eV and fast neutrons energies greater than 0.1 MeV there is a much higher displacement rate with fast neutrons.

The displaced atom is known as an interstitial atom, its vacant lattice site as a vacancy. The combination is known as a Frenkel pair. The atom displaced by the bombarding particle is known as the primary knock on atom (PKA). With fast neutrons as the bombarding particle, the PKA has sufficient energy to displace further lattice atoms causing a displacement cascade. The interaction of the interstitials and vacancies with themselves, each other and other defects in the metal results in swelling (5). When a Frenkel pair has been created the interstitial and vacancy have a variety of paths which they may follow.

Interstitials may interact with other interstitials to form two dimensional platelets which are areas of extra lattice planes known as dislocation loops. For a discussion of the possible configurations of multiple interstitials and further references on this subject the work of Miller (6) and colleagues should be consulted. Vacancies may aggregate into two dimensional vacancy loops which are areas of
missing lattice planes. Single vacancies should meet another vacancy before they reach point defect clusters and so form a divacancy (7) which then proceeds by thermal motion to the defect clusters. Under certain conditions vacancies may cluster into the three-dimensional morphology known as voids. Interstitials and vacancies may recombine annihilating each other or they may migrate to defect sinks present in the material. Defect sinks can be divided into three categories; (8) biased, unbiased and saturable sinks.

Unbiased sinks show no preference for capturing one type of defect over the other type i.e. the radius of capture is the same for interstitials and vacancies. The rate of absorption of defects is proportional to the product of the diffusion coefficient of the point defect and the difference in the concentration of the point defect in the bulk metal and at the sink surface. Included in this category are incoherent precipitates and grain boundaries. Voids may also be regarded as this type of sink (9).

Sessile dislocations, coherent precipitates and solute atoms are included in the saturable sinks category. When a dislocation line is pinned it will collect a finite number of interstitials around it, in the form of a dilute atmosphere. This will then act as a region of enhanced recombination and reduce the supply of vacancies to the void.

Any dislocation in the solid exhibits a preferential attraction for interstitials compared with vacancies. This bias is due to the non-random diffusion of point defects to dislocations caused by the strain field arising from the dislocation point defect interaction
Following Heald [10] the flux of either species to a dislocation is given by:

$$J + D \frac{CV}{kT} \text{flux} (r, \Theta) \quad 1.2$$

where $C$ is the concentration of the point defect

$D$ is the diffusion coefficient of the point defect

$V, k, T$ have the usual meaning

$r, \Theta$ are the polar coordinates of the point defect with respect to the dislocation.

$E_{\text{flux}} (r, \Theta)$ is given by Heald in the form:

$$E_{\text{flux}} (r, \Theta) = \frac{(1 + \nu) b \Delta V \sin \Theta}{3(1 - \nu) \pi r} \quad 1.3$$

for an edge dislocation in an isotropic material. In equation 1.3 the parameters have the following meaning:

$v$ is Poisson's ratio

$\lambda$ is the shear modulus

$b$ is the Burgers vector of the dislocation

$\Delta V$ is the local volume change regarding the point defect as the centre of dilatation in a continuum.

$\Delta V_v$, the relaxation volume of a vacancy is much less than $\Delta V_x$, that of an interstitial point defect. Typical values are given by Heald with $\Delta V_v$ from 0.2 to 0.5 and $\Delta V_x$ from 1.0 to 2.0, where $\Omega$ is one atomic volume unit. Thus the second term in equation 1.3 is higher for interstitials than vacancies, resulting in a greater flux of interstitials than vacancies to the dislocations.
the edge component of the dislocation can climb as a result of absorbing a vacancy or an interstitial. This does not hold if the dislocation climb is impeded by, for example, a precipitate.

Dislocations can be divided into two types. These are the network dislocations present in the unirradiated metal and augmented by unfaulting of the Frank dislocation loops and dislocation loops formed by agglomeration of interstitials. Bullough and Newman (11) calculated the interaction between point defects and long straight dislocations. They also calculated that between point defects and dislocation loops and obtained different answers for the interactions. However loops are assumed to be similar to straight dislocations because the dislocation stress field only affects point defects within approximately 10b from the dislocation core (12). Most loops are significantly larger than this. The only difference between dislocations in isolated loops and those in networks resides in the differing rates of thermal emission of vacancies (8). These are different because while an interstitial loop must of necessity increase its line length during vacancy emission, a network dislocation need not do so. As the interaction energy is taken to be the same for dislocation loops and the network dislocations the bias for both situations is the same.

Void formation and growth require three conditions to be satisfied. There must be a supersaturation of vacancies, the vacancies must migrate to voids before being annihilated by interstitials and more vacancies than interstitials must arrive at the void nuclei. The type of defect structure resulting from irradiation depends markedly on the temperature at which the
irradiation is carried out. The three conditions necessary for void formation are satisfied in most metals in the temperature range from 0.3T<sub>m</sub> to 0.6T<sub>m</sub>, where T<sub>m</sub> is the absolute melting point.

Below this temperature range the vacancy mobility is relatively low. The migration energy for vacancies is about an order of magnitude higher than the interstitial migration energy e.g. in nickel E<sup>v</sup> = 1.44eV, E<sup>i</sup> = 0.15eV (6). The low vacancy mobility leads to a large concentration of single vacancies. This means that the more mobile interstitials have a higher chance of interacting with a vacancy and recombining than of migrating to a sink. Above the stated temperature range the irradiation produced vacancy concentration is less than the thermal concentration. Thus the necessary vacancy supersaturation condition is not satisfied. The third condition is satisfied if there exists a biased sink for point defects which absorbs more interstitials than vacancies. This results in a larger flux of vacancies than interstitials arriving at void nuclei. The temperature range 0.3T<sub>m</sub> to 0.6T<sub>m</sub> encompasses for most metals the temperature at which the structural core components in a reactor are required to operate.

Although it is accepted that dislocations exhibit a bias for interstitials the value of this bias is uncertain. A selection of bias calculation results is presented in table 1.2. Nichols (13) has recently reviewed this topic and concluded that the lack of reliable measurements of the bias factor hampers understanding of void swelling.
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<tr>
<th>Author</th>
<th>Bias/%</th>
<th>Material</th>
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<tr>
<td>R. Bullough, B. L. Eyre &amp; K. Krishnan</td>
<td>8</td>
<td>316 S.S.</td>
<td>Symp. on physics of radiation produced voids. Harwell Sept. '74</td>
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<td>M. J. Makin &amp; G. P. Walters</td>
<td>&gt;5</td>
<td>316 S.S.</td>
<td>as above</td>
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<td>R. Bullough &amp; A. D. Brailsford</td>
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<td>J. Nuc. Mat. 44 121 ('72)</td>
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<td>R. Bullough, B. L. Eyre &amp; R. C. Perrin</td>
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<td>6</td>
<td>Nickel</td>
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S.S. = Stainless Steel

Table 1.2 Bias factor calculation summary
The treatment of Bullough and co-workers (14) has been correctly
criticised by Nichols and also by Foreman (15). Foreman points out
that ignoring the angular variation of the interaction between a
point defect and a dislocation results in a large overestimate of the
bias value. If the 1% to 2% value of Bullough et al. was, as Foreman
suggests, a factor of three or four higher than if they had taken the
angular variation into account, then the true bias is less than 1%.
This is not a reasonable value as it would not account for the
observed growth rate. Heald (16) also criticises this treatment on
the grounds that it suggests a diffusion controlled mechanism which
should lead to spherical voids. The results shown in chapter 4
suggest another alternative to Heald's proposal of a surface reaction
controlled mechanism. Brailsford and Bullough (8) adjusted their
parameters to obtain a value of bias in agreement with the Bullough
et al. value previously discussed. As this value is erroneous the
Brailsford and Bullough treatment can not be reliable. Wiedersich
(17) also obtains a value of 1% which again falls within Foreman's
criticism.

Heald's treatment has also been criticised by Nichols. Heald
ignores the presence of recombination but this is included in the
analysis of White and Fisher (18). White and co-workers have also
modified Heald's treatment for large dislocation densities such that
the infinite value of bias predicted by Heald in this regime does not
occur. Nichols also criticises the Heald approximation of no defect
production within the capture radius of the dislocation.
Thus there is no completely satisfactory model of these complicated interactions. However in order to perform experiments a model must be assumed and as the model used by Heald has been used with some success by Fisher et. al. [9] it is this model which was used in the course of this project. Fisher and White [20,21] suggest that a more complete analysis of void growth experiments is required in order to resolve some of the uncertainties surrounding this phenomenon. In their report [9] they suggest a technique for further investigations which is described in section 3.1.

The time scale involved makes "in reactor" experiments very difficult due to the defect production rate which is given by:

$$\phi = \phi_c \sigma_d \bar{N}$$

where $\phi_c$ is the fast neutron flux (approx. $8 \times 10^5$ ncm$^{-2}$s$^{-1}$)

$\sigma_d$ is the primary knock on cross section (approx. $2 \times 10^{-24}$ cm$^2$)

$\bar{N}$ is the average number of surviving defect pairs per primary knock on. (approx. 100). (17)

Thus in a typical fast breeder the defect production rate or displacement rate is of the order of $10^{-6}$ displacements per atom per second (dpa s$^{-1}$). With typical planned reactor lifetimes of some years the fast neutron fluence is of the order of $10^{23}$ ncm$^{-2}$ to $10^{24}$ ncm$^{-2}$. At this fluence each atom in a typical core structural material has been displaced from its normal lattice position about one hundred times.
The period of years involved in attaining the goal fluence has lead to the development of techniques for simulating the effects of high fluence, fast neutron irradiation. Using high energy charged particles from powerful accelerators, such as the high voltage electron microscope (HVEM) or the variable energy cyclotron, the atom displacement rate can be increased by orders of magnitude relative to that in the reactor core. Thus the state of radiation damage in a material after residing in the reactor core for a few years can be approached in a few hours. The effects of such increases in damage rate are complex. Thus while these techniques do give guidance to the parameters involved they can not be thought of as accurately reproducing the reactor environment. They can be used in conjunction with reactor data to extrapolate the effects of swelling to higher doses than easily obtainable from a short term reactor experiment. Recent work on the correlation of particle and neutron irradiation has been carried out by Mayer (104).

1.2. Simulation in the HVEM.

Use of the HVEM to investigate and simulate radiation swelling is now a well established technique with much of the pioneering work having been done by Norris (22) and Makin (23). This project involves the use of the Hitachi 1MV electron microscope at Berkeley Nuclear Laboratory with the permission of the Central Electricity Generating Board. Laidler et. al. (24) recently reviewed the advantages and disadvantages of this simulation method. As this is such a well documented technique the merits of it will only be stated briefly. The reader is referred to the literature for a full discussion (3,24,25,26,27).
Advantages

1. A single defect population can be observed continuously thus eliminating specimen to specimen variability.

2. A damage rate of the order of $10^{-3}$ dpa s$^{-1}$ is available so experimental irradiations need only a few hours.

3. In an electron microscope the situation is not complicated by a displacement cascade. The energy transferred to a heavy nucleus by an electron scattered through an angle $\Theta$ is given by (29):

$$E_t = E_{\text{max}} \sin^2 \frac{\Theta}{2}$$

$E_{\text{max}}$, the maximum transferred energy, which occurs for a "head on" collision is:

$$E_{\text{max}} = \frac{2E}{M} \left( E + m_0 c^2 \right)$$

where $E$ is the kinetic energy of the incident electron

$M$ is the mass of the target atom

$m_0 c^2$ is the rest energy of the electron.

Using values for nickel being bombarded by 1MeV electrons the maximum energy transfer is of the order of 70eV. An energy of the order of 25eV is required to displace an atom from its lattice site. The number of defect pairs produced by each collision is given, for example by Kinchen and Pease (29) as:

$$N = 0.5 \left( 1 + \frac{E_{\text{max}}}{2E_d} \right)$$

This was used by Oen (28) to calculate that a 1MeV electron can only transfer enough energy to create a single Frenkel pair. The primary knock on atom does not have enough energy to cause further displacements. Thus there can not be a displacement cascade.
4. In a reactor it is almost impossible to isolate the individual effects of radiation damage and of helium and hydrogen production. Microscope specimens can be seeded with helium before being irradiated if so desired.

5. The temperature of the irradiation environment can be varied easily and accurately in the microscope.

Disadvantages

1. Use of the HVEM means using specimens of the order of 1 micron thickness so surface effects must be incorporated into the theory. The electron beam is fully focussed for irradiation experiments which means that very small volumes of material are irradiated with the same maximum flux. Typically the fully focussed beam which is Gaussian profile has a spot size at half height of 3.6 microns (109).

2. The fact that a displacement cascade does not occur is advantageous as the displacement dose is calculated without assuming a damage function. It also means however that the results obtained from the HVEM are difficult to relate to those obtained from reactor experiments.

3. The temperature range at which swelling occurs is shifted relative to that in neutron irradiated materials when using electrons as the irradiating particles. Typically the temperature at which maximum swelling occurs is of the order of 100°C higher in the microscope. The origin of this temperature shift lies in the increased damage rate which leads to an increased amount of mutual recombination which in turn can play an important part in defect loss
4. Beam heating problems arise in the area of specimens being irradiated but by correct choice of beam current these can be reduced (30).

5. Great care must be taken to ensure that the microscope atmosphere is similar in any comparative experiments. Buswell et.al. have demonstrated that swelling is affected by the microscope environment (31).

1.3. Void growth theory

The rate theory continuum (R.T.C.) model was originally developed by Harkness and Li (32,33) and by Wiedersich (17) and has since been modified by many workers including Heald (10). The idea in this model is to replace all the discrete sinks in the body, including the voids, by equivalent continuous distributions of sinks in a continuum. In this continuum the various sinks are assigned strengths which ensure that the flux of defects to such sinks will be as close as possible to the flux to the actual sinks in the real body. Thus all spatial variations in the steady-state interstitial and vacancy concentrations are removed and the production and annihilation rate balance equations reduce to a pair of quadratic equations.

This model has been extended recently by Fisher et.al.(9) to be applicable to the HVEM environment. The theory will not be repeated 'in toto' in this report. Instead the expressions will be presented and details can be found in the series of reports (18,9,34,35). The
equations describing the point defect production rate and annihilation rate balance in the R.T.C. model are:

\[
\dot{\phi} - D_v C_v \gamma_v - D_v C_v \gamma_v - \alpha C_v C_I = \frac{dC_v}{dt} \tag{1.8}
\]

\[
\dot{\phi} - D_I C_I \gamma_I - D_I C_I \gamma_I - \alpha C_v C_t = \frac{dC_I}{dt} \tag{1.9}
\]

where \( \phi \) is the defect production rate

\( C_\beta \) is the concentration of point defects of type \( \beta \)

\( D_\beta \) is the diffusion coefficient of point defects of type \( \beta \).

The terms \( \alpha C_v C_I \) represent defect loss through mutual recombination and \( D_\beta C_\beta \) represent the defect loss to the surface. The parameters \( k_v \) and \( k_I \) represent the mean free path of an interstitial or vacancy respectively in the presence of the various sinks.

\[
k_v^2 = \frac{Z_v}{\rho_d} + \frac{Y}{\rho_v} \tag{1.10}
\]

\[
k_I^2 = \frac{Z_I}{\rho_d} + \frac{Y}{\rho_I} \tag{1.11}
\]

where \( Z_\beta \) represent the dislocation sink strength for the defects

\( Y \) is the void sink strength for the point defects

\( \rho_v \) and \( \rho_d \) are the void and dislocation densities respectively.

The \( Z_\beta \)'s and \( Y \) can be physically interpreted as the radius around a sink within which any point defect will interact with that sink.

Assuming a steady state then \( \frac{dC_v}{dt} = 0 \) and \( \frac{dC_I}{dt} = 0 \) and (1.8) and (1.9) can be solved to give expressions for the void and dislocation loop growth rates, given by:
\[ \frac{dr_v}{dt} = \frac{Y \phi F(\eta) (Z_+ - Z_-)}{4\pi r_v^3 (k_v^3 + k_s^3)(k_v^3 + k_g^3)} \]  
\[ \frac{dr_l}{dt} = \frac{\phi F(\eta) (Z_+ - Z_-)(k_v^3 + k_s^3)}{b (k_v^3 + k_s^3)(k_v^3 + k_g^3)} \]

where \[ \eta = \frac{4 \alpha \phi}{D_v D_\perp (k_v^3 + k_s^3)(k_v^3 + k_g^3)} \]

\[ F(\eta) = \frac{\phi}{\eta} \left( \left( 1 + \eta \right)^{\gamma} - 1 \right) \]

\[ \beta \] is the Burgers vector of the dislocation

and \( r_v, r_l \) are the void and loop radii respectively.

The parameters involved are discussed in (a) to (e) below.

These equations include the effects of mutual recombination and are from Fisher et al. (9).

\[ (a) \] Y, the void sink strength:

This is usually assumed to be \( 4\pi r_v \). In the presence of internal sinks and recombination an approximate expression for the sink strength was found to be, using iterative techniques:

\[ Y = Y_\beta^0 \{1 + \frac{Y_\alpha^0}{4\pi} \left( k_\beta^3 + \frac{1}{X_v} + \frac{1}{r_v} \right) \} \]

with \[ Y_\beta^0 = \frac{4\pi r_v^3}{r_v + r^*} \]

where \( R_v \) is the void spacing

\( r_v \) is the void radius

\( r^* \) is the interface radius

\( X_v \) is the vacancy mean free path.

The interface radius, \( r^* \), is defined by Heald (16) as:
Voids of radius smaller than \( r^* \) have polyhedral morphologies and grow by an interface controlled mechanism which depends on the spreading of atomic ledges across the polygonal faces of the void. Voids of larger radius than \( r^* \) are claimed by Heald to be spherical and to grow by a diffusion controlled mechanism.

(b) The surface sink strength, \( k^2 \):

An approximate solution for the surface sink strength in the presence of internal sinks and mutual recombination has been given by White et al. (32):

\[
k^2_s = \frac{2k}{t} \left\{ \frac{\sinh(kA t/2)}{\cosh(kA t/2) - 1} \right\}
\]

where \( \left( k_{1/2} \right)^2 = \frac{kA}{F[\eta]} \)

\( F[\eta] \) contains only the losses to the fixed internal sinks, and \( t \) is the foil thickness.

(c) Dislocation sink strengths \( Z \):

These have been calculated numerically, including mutual recombination, to be of the form:

\[
Z_\beta = Z_\beta^0 \left\{ 1 + \frac{\ln \left( L_\beta / b \right)}{21 + 0.0167 R_d / b} \left( \frac{R_d}{X_v} - 1 \right) \right\}
\]

for \( \frac{R_d}{X_v} \leq 20 \)

where \( Z_\beta^0 = \frac{2\pi}{\ln \left( \frac{2R_d}{L_\beta} \right) - \eta} \) and \( X_v = \left( \frac{D_\beta D_\varphi}{\alpha^2} \right)^{1/4} \)
The $L_A$ are the capture radii for the point defects and are given by:

$$L_A = \frac{(1 + \nu) \mu b \Delta V_A}{(1 - \nu) kT X_v^3}$$  \hspace{1cm} \text{(1.18)}$$

where $\nu$ is Poisson's ratio

$\mu$ is the shear modulus

$\Delta V_A$ is the defect relaxation volume

$R_d$ is the dislocation spacing

and $X_v$ is the vacancy mean free path.

For $\left(\frac{R_d}{X_v}\right) \gg 20$ and $D_v \gg Z_A^3 \left(\frac{R_d}{X_v} = 20\right)$.

(d) Recombination coefficient $F[\eta]$:

This depends primarily on $\phi$, and $D_v$ the vacancy diffusivity, as equation 1.14 shows. $D_v$ depends on $E_m^V$, the vacancy migration energy as $D_v \propto \exp\left(-\frac{E_m^V}{kT}\right)$.

(e) The displacement rate:

The damage rate is dependent on the displacement cross section $\sigma_d$ (equation 1.4). This depends on the displacement energy $E_d$ and its variation with direction. An isotropic value of $E_d$ is normally assumed.

As stated the HVEM technique is superior to neutron and ion irradiation as the displacement event is uncomplicated by multidefect production, an uncertain amount of immediate recombination and spontaneous aggregation. The theory given in section 1.2 has
therefore been used in conjunction with high voltage electron microscope experiments to obtain a value for the interstitial bias in nickel.
CHAPTER TWO

IMAGE SIMULATION
Introduction

In chapter one of this report it has been stated that void swelling is an important phenomenon in reactor technology. The estimation of swelling, $S$, by microscopy involves the determination of void size and void density (36). If the mean void volume is given by $\overline{V}$ and $\rho_h$ is the void density then

$$S = \frac{\overline{V}}{\rho_h}$$

The measurement of void volume involves an assumed shape and is generally calculated from a linear measurement (say $p'$) where

$$\overline{V} = c_o p'^2$$

where $c_o$ is a constant.

Fisher and Buswell (36) list as sources of error:

(a) the selection of a suitable parameter $p'$
(b) the measurement of $p'$
(c) the value assumed for $c_o$
(d) the calculation of $\overline{V}$ from $c_o, p'$
(e) the measurement of the void density.

Fisher and Buswell then suggest a procedure to standardise void swelling measurements. The suggested procedure does not however include any consideration of the focussing conditions operating in the microscope. When cavities, either voids or bubbles, are imaged in a defocussed mode Fresnel fringe contrast is seen at the projected edges of the cavity. This is shown in figure 2.1. The measurement of the linear parameter $p'$ can vary depending on which set of fringes are used for measurement. Rühle (25) has stated that for large spherical cavities (greater than 0.4 of an extinction distance in diameter) the outer diameter of the first dark ring corresponds to the actual bubble size when imaged with $-0.6 \mu m$ of defocus. Sprague
Out-of-focus contrast calculations for large bubbles ($R_b = 0.4 t_x$, $t_x = 250 \lambda$). The foil thickness is $3.75t_x$ for $\xi = +6000 \lambda$ and $\xi = -6000 \lambda$. The reduced intensity $I/I_0$ ($I_0$ = background intensity) is plotted vs. $\rho = r/R_b$. The in-focus contrast ($w = 0$) is also represented.

Fig. 2.1 Profiles of a through focal series. (Rhüle (25))
et. al. (37) state that the most accurate measurement is obtained by using an overfocussed image and measuring the line between the outside bright fringe and the inside dark one. These conditions are both derived for spherical cavities. Voids are however rarely spherical (38); they tend to be cubic or octahedral with truncations. Faceted shapes are also exhibited by large irradiation produced bubbles which in bcc metals tend to be brick shaped (39). As with voids there are no conditions laid down in the literature to obtain the most accurate size measurements.

During the course of this work a computer program has been developed which remedies this situation by simulating the electron microscope image of cavities. The program will show a means of differentiating between voids and bubbles by means of their strain field contrast. A method of dealing with the strain field contrast of non-spherical cavities will also be discussed. This has been modelled as being spherically symmetric and has also been considered in the non-symmetric case. These models will, it is hoped eventually divulge information regarding the value of the overpressure in large, faceted bubbles.

2.1. Previous image simulation

In 1957 Whelan and Hirsch (40,41) first applied the dynamical theory of electron diffraction introduced by Heidenreich (42) to study the contrast of stacking faults. These first calculations ignored the effect of inelastic scattering of electrons in passage through the foils. This effect of anomalous absorption was included in the calculations of Hashimoto, Howie and Whelan (43). In 1961
Howie and Whelan [44] extended the theory to include contrast from defects with continually varying lattice displacements such as dislocation lines.

In these calculations Howie and Whelan used the column approximation introduced in their previous work (45). This approximation amounts to assuming that the intensity of the electron beam at any point on the lower surface of a foil can be attributed to a thin column of material above that point. No account is taken of the interaction between waves diffracted by neighbouring columns as the difficulties involved in the numerical solution are greatly increased [44]. Arguments in favour of the column approximation are given in reference 45 but the main justification for the use of this approximation are stated by Howie and Whelan to be the agreement between experimentally observed images and those derived theoretically.

In their paper Howie and Whelan [44] give solutions of the wave equations for the two-beam case but also derive a method of solution which allows several diffracted beams to be included. The solutions in the two-beam case can be written in the form:

\[
\frac{d\phi'}{dz} = -\frac{\pi}{\xi_0} \phi' + \pi \left\{ \frac{i}{\xi_0} - \frac{1}{\xi_1} \right\} \phi'_q \tag{2.1}
\]

\[
\frac{d\phi'_q}{dz} = \pi \left( \frac{i}{\xi_q} - \frac{1}{\xi'_q} \right) \phi'_o + \left( \frac{\pi}{\xi'_q} + 2\pi c (z + \beta') \right) \phi'_q \tag{2.2}
\]

where \( \phi'_o = \phi_o \exp(-i\pi z/\xi_o) \)

\( \phi'_q = \phi'_q \exp(-i\pi z/\xi'_q + 2\pi c g z + 2\pi i g^2) \)

\[ \beta' = \frac{d}{dz} \{ g.R(z) \} \]
\( \phi_o \) is the amplitude of the transmitted beam
\( \phi_d \) is the amplitude of the diffracted beam
\( R(z) \) is the displacement function due to the defect
\( s \) is the deviation from the Bragg reflecting position
\( \xi_{j_1j_2} \) are the absorption distances of the transmitted and diffracted beams respectively
\( \gamma_{j_1j_2} \) are the extinction distances of the transmitted and diffracted beams respectively.

The fact that the displacement field of a defect occurs directly in these equations has lead to the technique being an important tool in electron microscopy. The theory has been used extensively in the identification of defects, especially dislocations and various forms of stacking faults. Van Landuyt et al. [46] and Gevers et al. [47] are examples of groups which have applied the theory to identify types of stacking faults. Numerous determinations of Burgers' vectors of dislocation loops have been made using this approach. Silcock and Tunstall [48] and Tunstall and Goodhew [49] are examples of groups who have applied the theory to partial dislocations.

In 1963 Ashby and Brown [50] applied the dynamical theory to the case of spherically symmetric strain fields due to spherical inclusions and specifically to face centred cubic (fcc) cobalt precipitate in the fcc copper matrix. McIntyre and Brown [51] considered the strain contrast from centres of dilation whose strength is similar to that of voids in aluminium. Van Landuyt et al. [52] calculated the contrast from small voids using the two-beam and column approximations. They assumed that the cavities could be modelled as being disc shaped and that there was no surrounding
strain field. Thus the contrast arising was solely due to thickness contrast. Ingram [53] synthesised the work of McIntyre and Brown [51] with that of Van Landuyt et al. [52] and calculated the contrast of small spherical voids surrounded by a spherical strain field. The column and two-beam approximations were again used in this work. Sass et al. [54] incorporated the work of Mura [55] with the dynamical theory to calculate the diffraction contrast arising from cuboidal inclusions. They conclude that a cuboidal inclusion can be distinguished from a spherical one by a 'notch' in the line of no contrast. They also suggest that an asymmetrical image results when the operating reflection corresponds to a plane which is not a symmetry plane of the cube.

All the results mentioned previously were displayed as intensity profiles i.e. a plot of the intensity along a line traversing the defect. In order to make a comparison with an experimental image the observer had to mentally construct a three dimensional image from several profiles or produce a densitometer map [56].

Head [56] in 1967 suggested a method for displaying computed images of dislocations and stacking faults directly on computer output paper by arranging for the printer to produce a grey scale by overprinting the symbols. The numbers, representing the intensity at the bottom of the foil, from the solution of the Howie and Whelan equations [44] could be displayed as a symbol of a certain density. A two dimensional array of these symbols constructed the image. In the same paper Head produced a technique for greatly increasing the speed of calculating the image of defects whose displacement field is plane stress or strain in nature. A large amount of work was carried out by
the Australian group of Head, Humble, Clareborough and Morton in the late 1960's and early 1970's using these methods. References 57 to 62 are examples from this group.

The basis for the speed of Head's method is that the displacement field around some defects is such that the displacement along any line parallel to the defect is constant. A generalised cross-section can be thought of with fixed dislocation position and displacement field. The variable position of the foil surfaces delineates the different parts of the field encountered by the electron beam in passage through the foil. The intensity at the lower foil surface is calculated by integrating the Howie and Whelan equations down columns of the foil using the fourth order Runge-Kutta process (63,64). Thölén (65) in 1970 published a rapid method of obtaining contrast maps from defects. The restriction of plane stress or strain imposed by Head is relaxed by Tholen as the contrast is calculated separately for each column. This means that any lattice defect can be treated using Thölén's method. Calculating the contrast separately from each column by integration is a time consuming process so Thölén made use of the matrix method suggested by Howie and Whelan (44) and used by Gevers, Van Landuyt and Amelinckx (66).

The matrix method is shown schematically as figure 2.2. The Howie and Whelan equations have two independent solutions which can be given in an exact form for the case of a perfect crystal. The total wave is a linear combination of the two solutions which satisfy the correct boundary conditions at the top surface. The relation between the waves at the top and bottom of a foil of thickness, t, is given by Thölén in the form:
Figure 2.2 Matrix model for image simulation
where the matrix \((\Phi^0(z))\) is defined as the wave matrix.

This work follows the work of Van Landuyt et. al. (viz. figure 2.2) where transmission of the electron beam through the first part of the crystal of thickness \(z_1\) is represented by a matrix \(A_1\). The matrix \(V(z_2)\) takes into account the presence of a cavity of thickness \(z_2\), and finally \(A_3\) describes the transmission and scattering through the perfect part of the crystal of thickness \(z_3\).

For a column passing through the cavity the amplitudes \(\Phi_0^V\) and \(\Phi_3^V\) of the transmitted and scattered wave at the exit surface of the foil are given by:

\[
\begin{pmatrix}
\Phi_0^V \\
\Phi_3^V
\end{pmatrix} = A_3 V(z_2) A_1 \begin{pmatrix} 1 \\ 0 \end{pmatrix}
\]

The matrix \(V(z_2)\) incorporates the phase change which the beam undergoes in passage through a hole of thickness \((z_2)\). This phase shift is due to the difference of the mean inner potential \(V_0\) in the cavity and in the perfect crystal. Rühle (25) has calculated that the phase difference between a beam passing through a cavity of thickness \(z_2\) and a beam passing only through perfect crystal is given by:

\[
\exp\left(-2\pi i \xi'(z_2)\right)
\]

where \(\xi'\) is given by:

\[
\xi = \frac{e V_0}{2 E \lambda}
\]

\(e\) is the electronic charge

\(V_0\) is the mean inner potential

\(E\) is the total energy of the electron

\(\lambda\) is the wavelength of the electrons.
For a column of crystal which does not pass through the cavity, the amplitude of the transmitted and scattered waves is given by:

\[
\begin{pmatrix}
\phi_t^p \\
\phi_t^s
\end{pmatrix} = A(t)
\begin{pmatrix}
1 \\
0
\end{pmatrix}
\]

where \( t \) is the foil thickness equal to \( z_1 + z_2 + z_3 \).

The intensities of the transmitted and scattered beams can easily be calculated from the amplitudes at the lower foil surface. The amount of normal absorption suffered by the electron beam depends on the thickness of crystal through which the beam travels. Thus the level of normal absorption will be different for columns passing through the cavity and columns passing only through perfect crystal (52). The intensities must be multiplied by a factor of

\[
\exp \left( -\frac{y}{\lambda} \right)
\]

where \( \lambda \) is the normal absorption coefficient and \( t_\lambda \) is the amount of crystal the beam travels through in the column under consideration.

This point will be further discussed in section 5.1.

In the case studied by Van Landuyt et al. (52) of strain free, disc shaped voids the matrices \( A_1 \) and \( A_2 \) and \( V(z_2) \) can easily be calculated. However when considering cavities with a surrounding strain field the assumption of perfect crystal outside the cavity cannot be made. Tholen surmounts this problem by dividing each column into a number (n) of crystallites each of thickness \( \Delta z \) and calculating the scattering matrix corresponding to each point in the foil. This is shown schematically in figure 2.3. As the Howie and Whelan equations are linear the matrix elements are not functions of \( z \) but only of \( \Delta z \) and \( w \). The individual elements are obtained by calculating the wave fields at the bottom of a crystal slab of
Wave matrices

Top surface

\[ \emptyset \]

\[ A_1 \emptyset \]

\[ A_2 A_1 \emptyset \]

\[ A_3 A_2 A_1 \emptyset \]

\[ A_n A_m A_l \emptyset \]

\[ \emptyset_c \]

\[ \emptyset_3 \]

Displacement field due to defect

Line defect

Lower surface

Figure 2.3  Thölén's multislice matrix method
thickness Δz with the starting values \( \phi_0 = 1, \phi_2 = 0 \) and \( \phi_0 = 0, \phi_2 = 1 \) respectively.

\[
a_{11} = \frac{-\left(\chi_1 + \frac{\pi \xi_2}{\xi_0}\right) \exp(\gamma_1 \Delta z)}{\gamma_1 - \gamma_2} + \frac{\left(\chi_1 + \frac{\pi \xi_2}{\xi_0}\right) \exp(\gamma_2 \Delta z)}{\gamma_1 - \gamma_2} \quad 2.8
\]

\[
a_{21} = a_{11} = \frac{\pi \left(\chi_1 - \frac{\xi_2}{\xi_0}\right) \left(\exp(\gamma_1 \Delta z) - \exp(\gamma_2 \Delta z)\right)}{\gamma_1 - \gamma_2} \quad 2.9
\]

\[
a_{22} = \frac{\left(\chi_1 + \frac{\pi \xi_2}{\xi_0}\right) \exp(\gamma_1 \Delta z) - \left(\chi_2 + \frac{\pi \xi_2}{\xi_0}\right) \exp(\gamma_2 \Delta z)}{\gamma_1 - \gamma_2} \quad 2.10
\]

where

\[
\gamma_{1,2} = -\pi \frac{\xi_2}{\xi_0} + \ln \sum \left\{ \omega \pm \left(\omega^2 + 1 - \frac{\xi_2^2}{\xi_0^2} + 2i \frac{\xi_2}{\xi_0}\right)^{1/2} \right\} \quad 2.11
\]

In the relationships all distances are given in terms of extinction distances, \( w = s \xi_1 / \xi_0 \), and normal absorption is included. The effective deviation parameter \( w_{\text{eff}} = w + \xi \beta \) is calculated in the first slice and this corresponds to a particular matrix \( A_1(w_{\text{eff}}, \Delta z) \). The wave matrix at the top surface is multiplied by \( A_1 \) and a new wave matrix is obtained which is then the wave matrix at the top of the second slice. In slice two a different matrix \( A_2 \) is needed as \( w_{\text{eff}} \) is changing and \( A_2 \) is then used to calculate the wave matrix at the top of the third slice and so on down the column. Starting with \( \phi_0(0) = 1, \phi_2(0) = 0 \), the bright and dark field amplitudes at the bottom of the foil are calculated from:
Thölen points out that for a foil of four extinction distances thick divided into 80 slices a typical contrast computation will involve calculation of the matrix elements at about $10^5$ points. As many of the matrices will be almost identical Thölen calculates and stores a library of matrices corresponding to different values of $\omega$. He then states that as $\omega$ is continuously changing a small range of $\omega$ values, $\omega + \Delta \omega$, can be assigned to the matrix value calculated for $\omega$. The computations carried out in this project are based on Thölen's method.

If the images being considered are in the 'in focus' condition the phase information at the lower foil surface is lost when the intensities are calculated. The phase factor at the lower foil surface becomes important when imaging in the defocussed mode. Rühle (25) in 1971 calculated the contrast from spherical voids and bubbles taking account of the focussing conditions operating in the electron microscope. He used the two-beam approximation and the column approximation in the solution of the Howie and Whelan dynamical equations. Figure 2.4 is a schematic representation of the method of calculating the image at a defocussed plane from that at the 'in focus' plane.

The method consists of first calculating the Fourier transform of the amplitude field existing at the 'in focus' image plane. A transfer function must then be calculated to account for the phase shift occurring in the beam while propagating from the 'in focus' to
Foil top $Z = -t/2$

Defect

In focus plane

Foil bottom $Z = t/2$

Transfer function 'T' accounts for phase shift in this region

Defocussed plane

$Z = \Delta f + t/2$

Figure 2.4 Image planes
the defocussed plane. This transfer function includes the lens aberrations and coherence effects. The amplitude of the Fourier partial waves is then multiplied by the transfer function and an inverse Fourier transformation performed to obtain the amplitude existing at the defocussed image plane. In the work of Rühle (25) and Rühle and Wilkens (67) the cavities are assumed to have rotational symmetry with respect to the electron beam direction as this allows an analytical solution to be obtained for the defocussed image. The technique will be discussed more fully in section 5.2.

In their work on the image of spherical cavities which are not surrounded by a strain field Rühle and Wilkens (67) extended the work of Van Landuyt et. al. (52) to a systematic N-beam case. Systematic reflections will always be present to some extent even when diffraction conditions are such that a strong reflection is operating. The number of systematic reflections increases with the atomic number of the specimen and accelerating voltage used in the microscope (68).

The effect of using only two beams to construct the image is to underestimate the magnitude of the extinction distance. Evaluations of the errors involved were given by Howie and Basinski (69) and by Serneels and Gevers (70). For the case of spherically symmetric coherency strains, considered by Ashby and Brown (50), Howie and Basinski conclude that the error, due to using the two beam approximation is small. Edington (71) summarised the effect of including many beams in the image calculations of various types of defects. It can be concluded from the literature that use of the two-beam approximation is justified if care is taken over the
diffraction conditions to be modelled. In particular the specimen must be tilted to nearly two beam conditions such that the first order reflections are used and the deviation from the Bragg position is small. This implies dynamical diffraction conditions with strong $g$ and small $s$. The specimen tilt must also be such that no non-systematic reflections occur.

Images of defects with the surrounding strain field not in contrast are usually obtained in kinematical diffraction conditions, i.e. weak $g$ and large $s$. Many beam theory may give a more accurate absolute intensity map for these images. It will be shown however that the position of the Fresnel fringes surrounding a defect from which measurements are made are independent of extinction distance. This is the parameter effected by many beams being considered so the measurements should not be changed by using only two beams in the calculations.

2.2. Strain field calculations

The ultimate aim of this project is to obtain information on the strain field surrounding voids and bubbles. Strain fields are imaged using two-beam dynamical diffraction conditions so use of the two-beam approximation is valid.

The justification given by Van Landuyt et. al. (52) for ignoring the strain field is that the vapour pressure inside a bubble can exactly balance the surface tension forces, leaving the surrounding matrix strain free. It has been pointed out that this is not the case for a gas filled bubble in equilibrium with the surrounding matrix (72,73).
Lidiard and Nelson (72) showed that in a solid in true thermal equilibrium where mass transport by vacancies or other defect mechanisms is possible the equilibrium conditions are:

\[ P = \frac{2\gamma}{r_c} \quad \text{instead of} \quad P = \frac{2\sigma}{r_c} \quad 2.13 \]

Where \( \gamma \) is the surface energy
- \( \sigma \) is the surface tension (or surface stress)
- \( r_c \) is the radius of the cavity
- \( P \) is the gas pressure.

The surface tension, \( \sigma \), and the surface energy, \( \gamma \), are not equal for solids; in fact they are related by the equation (73):

\[ \sigma = \gamma + A \left( \frac{\partial \gamma}{\partial A} \right) \quad 2.14 \]

In equation 2.14 the differentiation indicates a change in area \( A \) of the bubble under conditions in which the number of surface atoms remain constant. The existence of a surface tension different from the free surface energy implies that a strain field exists around a bubble in thermodynamic equilibrium the strength of which is proportional to \( \gamma - \sigma \).

Previous calculations of contrast maps including the strain field of voids and bubbles have always assumed spherical symmetry for the strain field (53,25) and have obtained black/white lobe contrast as is the case with inclusions (74). This is sketched in figure 2.5 which is from Rühle (25). It is interesting to note that in the literature it is stated that the nature of a defect cluster may be decided from the direction of the black/white contrast, and the
defect depth is in extinction distance units from the nearest surface.

l = direction of black-white lobe contrast

Figure 2.5 Schematic of black/white contrast from small defects of vacancy type (from Ruhle (25)).
direction of the operating reflection. The sign of g.l depends on whether the defect is a vacancy or interstitial cluster as well as on the distance of the defect from the nearest foil surface. This would suggest that a similar technique could be used to differentiate between small voids and bubbles whose depth could be measured. The direction of the displacement in the surrounding matrix should be opposite when comparing that due to an overpressurised bubble to that due to a void. This will be investigated in section 6.2.

Although workers in the image simulation field have always assumed a spherical strain field and a spherical morphology for voids and bubbles, there have been calculations made with regard to the void lattice structure which do not make this assumption (75,76,77,78, 79). In their calculations these authors have also accepted that large voids are highly faceted unlike the spherical models used in the image simulation field. Using the more realistic shape for a cavity leads to problems when calculating the displacement field. Eshelby's (80) discussion of the general ellipsoidal inclusion implies that a spherical void can be simulated by an appropriate spherical inclusion. Unfortunately this simple model does not apply to a highly faceted void.

The approach used by the workers (75 to 79) is based on the fact that the displacement at any point in a matrix is related, via a Green's function, to the force acting at any other point (81). The displacement is given by:
\[ U^i_i (r) = G_{ij} \{ r - r' \} F^j_j (r) \]

where \( U^i_i (r) \) is the displacement at point \( r \), in direction \( i \).

\( F^j_j (r') \) is the force in direction \( j \) centred on \( r' \).

\( G_{ij} \) is the Green's function.

Thus the displacement at point \( r \), in direction \( i \), due to a force \( F \) centred on point \( r' \) and in direction \( j \), is given by the Green's function \( G_{ij} \) times that force. Bacon et al. (82) recently published a useful review of the theory of lattice defects in which the Green's function method is explained.

In order to use this method some system of forces must be derived to represent the void. Stoneham (76) used radial forces applied at various positions on a sphere centered on the void. He tried four sets of forces; simple cubic acting along \( <100> \); body centered cubic acting along \( <111> \); face centered cubic acting along \( <110> \) and also a spherical force. In this paper he does not give a meaningful assessment as to which forces may best represent the void. Malén and Bullough (75) represent the void by a spatial distribution of body force with body centered cubic symmetry. They achieved this by imposing eight equal point forces an equal distance from the origin and each acting towards the origin along a \( <111> \). These authors admit that their 'star of forces' representation of a void is rather crude but point out that a precise model of a faceted void would be extremely complicated.

Faivre (83) used the method suggested by Eshelby (80) to calculate the elastic field due to a perfectly coherent precipitate having the form of a rectangular parallelepiped. In this work he assumed that
the material was elastically isotropic and homogeneous. It is implicit in this method that the elastic constants of the material in the inclusion are the same as those in the matrix material. Melander (84) considers anisotropy in his calculations of the strong and weak beam contrast from cuboidal inclusions. He again assumes that the elastic constants in the defect are equal to those in the matrix. When modelling the strain field due to a cavity this approximation amounts to ignoring the missing material within the cavity. The errors involved in using this method have been discussed by Meissner (111). Further discussion of this point and a new technique for modelling large faceted cavities with non-spherically symmetric strain fields will be given in section 5.4 of this report.

There are many variables involved in image simulation and before moving to the details of the techniques involved it may be useful to list the parameters and their effect on the image, see also Bell et al. (85).

2.3. Parameters involved in image simulation

(a) Foil thickness:

The level of the background intensity depends on the relationship between the foil thickness and the extinction distance of the operating reflection. If the foil is an integral number of extinction distances thick, the bright field background is a maximum and the dark field background is a minimum. If the foil is an odd number of half extinction distances thick, the bright field intensity is a minimum and the dark field intensity is a maximum.
(b) Cavity dimensions:

The effect of the size of the cavity depends on the thickness of the foil in which the cavity is situated. Small bubbles, of size less than about one quarter of an extinction distance will be invisible in thick foils if imaged in focus. This is because the cavity will act as a pure phase object and thus has no effect on the intensity at the lower foil surface. The cavity will however be visible if imaged in the defocused mode. Large cavities in thick foils exhibit contrast mainly due to normal absorption, the contrast resulting from the difference in foil thickness traversed by the electron beams which pass through the cavity and those that do not. The cavity need not appear brighter than the background; this depends on the relationship between the cavity dimension parallel to the electron beam, the foil thickness and the extinction distance of the operating reflection.

(c) Defect depth:

When the diffraction conditions are such that the deviation from the Bragg condition is zero the depth of the defect, for large cavities does not effect the image. If the cavity is small enough such that there is a significant strain contribution to the contrast the depth of the defect becomes important. The contrast behaviour becomes more complex for diffraction conditions far from the Bragg position as the sign and magnitude of the contrast depend on the position of the cavity.
[d] Extinction distance:

Changing the extinction distance of the operating reflection is effectively altering the foil thickness. The effective extinction distance is given by:

\[ \xi' = \frac{\xi}{1 + s^2 \xi^2} \]

where \( \xi \) is the extinction distance at the Bragg angle 
\( s \) is the deviation parameter.

Thus the effective extinction distance can be changed by varying the value of \( s \) or by changing the operating reflection.

[e] Diffraction vector \( q \):

Lower order reflections have lower extinction distances and are normally used as they are more readily available. For strain contrast work however it is preferable to use higher order reflections having longer extinction distances.

[f] Anomalous absorption parameter:

The absorption distance is given by the equation:

\[ \xi' = \xi^0 (1 + \frac{\xi^2}{\xi^0 q^2}) \]

where \( \xi^0 \) is the minimum absorption distance.

Thus the absorption distance is a function of extinction distance and of the deviation parameter. It is generally assumed that the absorption distance is much larger than the extinction distance with
The approximate relationship:

\[
\frac{\delta}{\lambda} = 0.10
\]

\[
\frac{2\delta}{\lambda} = 2.18
\]

(g) Deviation parameter \( \delta \): 

The Bragg position, where \( \delta = 0 \), represents the position that approaches the optimum for obtaining strain contrast images. This is because tilting or rotation of the lattice near a defect can cause either a local increase or decrease in the transmitted intensity from the background in a perfect crystal. If the background is at a maximum, strain images are strongest with \( \delta \neq 0 \) as this allows the intensity to vary up or down. Exactly at the Bragg position, the intensity could not increase. The deviation parameter plays an important role in determining, for defects distributed throughout a foil, which defects display most contrast.

(h) Focussing conditions:

The apparent shape and size of a cavity is greatly influenced by the distance of the focussing plane of the objective lens from the lower foil surface. Cavities which are almost invisible when imaged in focus exhibit strong contrast in either over or underfocussed images.
CHAPTER THREE

EXPERIMENTAL
Introduction

The experimental work in this project can be split into two main areas. One was to produce and image voids in a high voltage electron microscope with a view to obtaining a value for the interstitial bias. The other was to obtain helium bubble images in well defined diffraction conditions using standard electron microscopes to match to those obtained from the computations. The comparison of the experimental and theoretical images may shed some light on the approximate value of the pressure of gas in a bubble.

3.1. Defect Bias

The experimental technique used to study the defect bias in nickel in this project follows directly that used by Fisher et. al. (93 in their analysis of copper. Equations 1.12 and 1.13 show that the magnitude of the function $Z_I - Z_v$ is of importance in the study of swelling. Unfortunately neither the magnitude of the $Z_I$'s themselves nor of the bias are well established. The bias is given by:

$$ B = \frac{Z_I - Z_v}{Z_I} $$  \hspace{1cm} 3.1

The expressions given for $Z_I$ are derived from calculation of the dislocation to point defect interactions due to the stress field of the dislocation and the point defect relaxation volume $AV_j$. Various workers have calculated the vacancy formation volume in nickel and have arrived at similar results [6]. The results suggest a vacancy relaxation volume of approximately $0.19\alpha$. With regard to the interstitial relaxation volume the situation is less clear as
calculated values vary by a factor of two \(6\).

Thus the determination of \(Z^V - Z^L\) and \(E^V\) are the major problems set by the parameters in equations 1.12 to 1.18. Equation 1.13 suggests that the results of a dislocation loop growth experiment at one temperature would yield a set of ambiguous values for \((B, E^V_m)\) where \(B\) represents the interstitial bias. Repeating the experiment at different temperatures and plotting the values of bias against those values of \(E^V_m\) which yield the observed growth rate at each temperature should yield unique values of bias and \(E^V_m\) satisfying the results at all temperatures. If \((Z^V - Z^L)\) and \(E^V_m\) can be determined then a comparison of loop and void growth in the early stages of irradiation will yield information on the void sink strength. Using equations 1.12 and 1.13 the following relationship between the void and loop growth rate can be obtained:

\[
\frac{\dot{r}_L}{\dot{r}_V} = \frac{(Y^b + k_s^2) 4\pi r^2_V}{b Y^d} \frac{k_s^2 4\pi r^2_V}{b Y^d}
\]

where \(\dot{r}_V\) is the void growth rate

and \(\dot{r}_L\) is the loop growth rate.

The HVEM is the most suitable instrument to carry out these experiments as temperature can be varied accurately and easily and void and loop growth data can be obtained. In equation 1.13 the observable parameters are the void and loop radii, the void and loop densities, the dislocation density and the foil thickness. The methods used for measuring these factors are given in section 3.1.1.
3.1.1. Measurement techniques

(a) Dislocation density:

The line dislocation density was measured using the method of Ham (86). A set of random lines with a total length, \( L \), was drawn in area, \( A \), on the micrograph print. The number of intersections, \( N \), which the grid lines made with the dislocations was counted. The total projected length, \( R_p \), of dislocation line in the given area, \( A \), on a micrograph is given by:

\[
R_p = \frac{\Pi NA}{2L}
\]

The dislocation density is given by:

\[
\rho_d = \frac{4R_p}{At\Pi}
\]

where \( t \) is the foil thickness.

From equations 3.2 and 3.3 it can be seen that

\[
\rho_d = \frac{2N}{Lt}
\]

(b) Foil thickness:

The foil thickness was either estimated by counting the number of fringes in an adjacent grain boundary (87) or by stereomicroscopy if suitable surface irregularities were present. In the former case the foil thickness, \( t \), is given approximately by:
\[
t = n \frac{\xi}{j_d}\]

where \( n \) is the number of fringes occurring

and \( \xi \) is the effective extinction distance corresponding to

the diffraction vector being used.

In order for the effective extinction distance to be as close as
possible to the tabulated value of extinction distance the value of
the deviation parameter, \( s \), should be a minimum. Thus the diffraction
conditions were such that \( s \neq 0 \) when the number of fringes was
counted.

(c) Stereomicroscopy

Stereomicroscopy has been used in measuring the dislocation
loop density and the void density. It has also been used in some
cases to measure the foil thickness. The theory of the method and the
errors involved have been discussed by Nankivell [88]. Hudson et al.
[89] have given the optimum tilt angles to use as a function of foil
thickness and magnification.

The principle of this method is that two micrographs are taken
of the same area of the specimen at the same magnification but at
different angles of tilt. The projection of the position of an
artifact within the foil will depend on the depth of the artifact. The
parallax can then be measured between pairs of points. This can be
measured either with a stereoviewer, or, if individual defects can be
easily identified a ruler can be used. The author has used an \((x,y)\)
travelling microscope which has been modified to take the readings
semi-automatically, at Berkeley Nuclear Laboratory. The voltage
analogues of the readings are taken on a digital voltmeter which is interfaced to a compatible desk computer which produces the required heights (36). The parallax, \( \Delta Y \), is related to the angle of tilt, \( \theta \), between the two micrographs, the magnification, \( M \), and the vertical separation, \( \Delta h \), of the two points by the relation:

\[
\Delta Y = 2\Delta h M \sin \frac{\theta}{2}
\]

\[ 3.7 \]

(d) Void density

In order to determine the void density it is not sufficient to count the number of voids present in an area on the micrograph and calculate the volume assuming that the voids are distributed throughout the foil. The presence of a layer close to each surface denuded of voids would make void density measurements of this form spurious. Using stereomicroscopy of an area containing a reasonable number of voids the width of the void layer, \( t_d \), can be determined. The void density \( \rho \), in an area, \( A \), containing \( n \) voids, can then be calculated from the relation:

\[
\rho = \frac{n}{A t_d}
\]

\[ 3.8 \]

(e) Dislocation loop density

When calculating the dislocation loop density it must be remembered that some proportion of loops will be invisible depending on the diffraction vector being used. This is because of the \( a,b \) invisibility criterion. Hirsch et. al. (87) list the proportion of
invisible loops as a function of the operating diffraction conditions. If the number of loops present in an area \( A \) is, after adjustment to include invisible loops, \( n_d \), then the loop density, \( N_L \), is given by:

\[
N_L = \frac{n_d}{A_t \Delta}
\]

3.1.2. Specimen preparation

Nickel was received in the form of 50-Hm sheets of 99.995% purity. Discs of 3mm diameter were punched from this and annealed for one hour at 760°C. The discs were then thinned using a Struers Tenupol twin-jet electro-polisher. The electrolyte used was 80% ethanol, 10% glycerol and 10% perchloric acid at a temperature of approximately 10°C. Methanol was used to rinse the discs which were then dried using lint-free tissue paper.
The specimen discs were then ready to be irradiated in the 1 MeV electron microscope to study the void growth.

3.2. Helium bubble imaging

Irradiation produced helium bubbles in niobium/1% zirconium and vanadium were imaged using the 100B and 200CX JOEL electron microscopes in the Microstructural Studies Unit at Surrey University. The samples were received in 3mm disc form with all the necessary preparation having been carried out by S.K.Tyler. The details of the preparation technique are given by Tyler in reference 39 but a brief summary may be useful. Helium gas was introduced into the 25μm foils by sequential implantation at 220, 140, 100 and 50keV nominally at room temperature. This treatment resulted in a uniform helium concentration of about 1at% at all depths between 2μm and 8μm from the implanted surface. The specimens were then annealed and then electropolished to a suitable thickness. The Vanadium specimens studied in this work were annealed at 1150°C for 8 hours and the niobium/1% zirconium specimens were annealed at 1250°C for either 2 or 8 hours.

The bubble dimensions were determined by measuring the length of the projected edges of the bubble with sufficient defocus to ensure clear edge images. Bubble images were usually obtained with about 120K magnification on the negative and with about 8μm of defocus. The defocus calibration for the microscopes was obtained from the manufacturer.
CHAPTER FOUR

DEFECT BIAS RESULTS AND DISCUSSION
4.1. Defect Bias

Quantitative analysis of the experimental data was performed using the simulation program of Fisher et. al.(9). A flow chart of this program is reproduced as figure 4.1. It was stated in section 3.1 that values must be found for \( E_m^v \) and \( \Delta V_T \). In the program the parameters are varied and the pairs of values which give the best match between the theoretical and the observed void growth rate noted.

The simulation program is based on equations 1.8 to 1.18. From equation 1.12 to 1.13 growth rates are calculated for a small time increment and the sink strengths are then updated at the end of this increment. New defect concentrations are calculated which are used to calculate new growth rates for the next time increment. Any discussion of the nucleation problem is avoided in the program by starting with the observed density of dislocation loops at an arbitrary small size and introducing the voids at the dose and size with which they are first observed. In test calculations Fisher et. al. found that the loop growth rate was insensitive to starting size and chose an initial loop radius of 50A. It is reasonable to introduce the observed loop density immediately as the loop nucleation time is effectively zero. If the loop nucleation time was measurably non-zero then the calculations could be started at this later time.
Set dose = 0
Set initial loop radius = 20xb
N = 1
Calculate dislocation sink strengths \( Z_j, Z_v \)

- inter - dislocation spacing > vacancy mean free path
  Yes
  Recalculate \( Z_j, Z_v \) with mutual recombination
  No

Set dose interval
Calculate total sink strengths \( K_j^2 + K_v^2 \) - for dislocations, voids (with interface controlled growth) and the surfaces
Correct \( K_j^2 \) and \( K_v^2 \) for second order effects
Calculate defect concentrations

- Has maximum dislocation density already been reached
  Yes
  Dislocation density set to reduced value
  No

- Loop radius > saturation radius
  Yes
  Dislocation density set to maximum
  Saturation dose recorded
  No

Increase average loop radius and calculate equivalent dislocation density
Calculate void growth for those voids nucleated in the first \( N \) steps

Increase dose by dose interval

- Is dose = No
  Yes
  Calculate volume swelling
  Calculate average void radius

N = N + 1

- Is \( N > \) maximum dose
  No
  Calculate bias
  Stop
  Yes
  Nucleate a further density of voids, \( D(n) - D(n-1) \), of initial void radius = 4xb
Set dose = 0

FIGURE 4.1 The Computer Program Flow Diagram.
From the loop growth rate and measured loop density a saturation radius and saturation time can be determined at which loop interaction begins. At times greater than this the dislocation network density was introduced into the program. Figure 4.2 shows a typical irradiation sequence with loops interacting to form a network and then voids appearing and subsequently growing. This irradiation took place at a temperature of 485°C and will be used as an example of evaluation of the experimental data. The defect structure present as a result of irradiation at 420°C is shown as figure 4.3. In this figure the diffraction conditions are adjusted to show the grain boundary fringes.

4.1.1. Derivation of data to be input to the program

The electron flux was measured using a Faraday cage and used to calculate the dose and damage rate. A typical Faraday cage reading was 7mV recorded through a 1MΩ resistor such that the current was $7 \times 10^{-9}$ amps. From the area of the aperture, of diameter 0.325 mm, the electron flux was calculated to be approximately $5 \times 10^{-23}$ em$^{-2}$ s$^{-1}$. The displacement rate could then be calculated using equation 1.4 to be about $2 \times 10^{-3}$ dpa s$^{-1}$ using a displacement cross section of 40 barns (22).

Measurement of the mean loop radii was made by measuring the radii of about one hundred loops in each of the micrographs. The loop density was calculated by counting the number of loops occurring in a given volume of the specimen allowing for invisibility criteria. In this case there 145 loops visible in a 10cm * 10cm area on the print using $g = [2 2 0]$. Thus one sixth of the loops are invisible as they
Nickel temp. = 485°C, mag. = 60K

a. 0.27 d.p.a.  
b. 0.60 d.p.a.

Dislocation loop regime  
loops beginning to interact

c. 2.10 d.p.a.  
d. 3.56 d.p.a.

voids are now visible

e. 4.05 d.p.a.  
f. 6.46 d.p.a.

Figure 4.2 Irradiation sequence
Nickel temp. = 420°C, mag. = 60K

Dose = 17 d.p.a.

Figure 4.3 Grain boundary fringes
have a Burgers vector of type a/2 [1 1 0]. Thus the number of loops in this area is 174.

From stereomicroscopy of the void layer the parallax between the top and bottom void is measured at 5.5 mm which corresponds to a vertical height difference of 3300 Å between the lowest and highest void. The magnification on the print is 60K which means that there are 174 loops in a volume of \(9.2 \times 10^{-13} \text{ cm}^3\) which in turn leads to a loop density of \(1.85 \times 10^{14}\) loops \(\text{cm}^{-3}\). The foil thickness was estimated at 4300 Å as five thickness fringes could be seen using \(g = [3 1 1]\) for which the extinction distance at 1 MeV is 850 Å. The dislocation density was measured using the method of Ham explained in section 3.1.1. In the dislocation loop regime the dislocation line density is calculated using the formula:

\[
\phi = 2\pi r^2 N_L
\]

where \(r^2\) is the mean loop radii.

The mean void radius was obtained by measuring of the order of one hundred voids in each micrograph. The void density was calculated in a similar way to the dislocation loop density.

The loop interaction radius is given by:

\[
R_I = \frac{3\sqrt{3}}{4\pi N_L}
\]

where \(N_L\) is the loop density. In this case \(N_L = 1.85 \times 10^{14}\) loops \(\text{cm}^{-3}\) and therefore \(R_I = 1800\) Å.

The void radii and density data are tabulated in table 4.1 and plotted in figures 4.4 and 4.5 against dose. The dislocation loop radii and dislocation line density data are plotted in figure 4.4 and
Table 4.1 Void growth data at 485°C

<table>
<thead>
<tr>
<th>Dose /d.p.a</th>
<th>Void density / * 10^5 cm^-3</th>
<th>Void radius / A</th>
</tr>
</thead>
<tbody>
<tr>
<td>2.14</td>
<td>1.3</td>
<td>56</td>
</tr>
<tr>
<td>3.55</td>
<td>0.91</td>
<td>74</td>
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<td>1.08</td>
<td>85</td>
</tr>
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<td>6.46</td>
<td>0.97</td>
<td>105</td>
</tr>
<tr>
<td>7.27</td>
<td></td>
<td>109</td>
</tr>
</tbody>
</table>

Table 4.2 Dislocation loop radii and line density data, irradiation temperature = 485°C

<table>
<thead>
<tr>
<th>Mean Loop radii / A</th>
<th>Dose / d.p.a</th>
<th>Dislocation line density / *10^3 cm cm^-3</th>
</tr>
</thead>
<tbody>
<tr>
<td>370</td>
<td>0.18</td>
<td>---</td>
</tr>
<tr>
<td>445</td>
<td>0.27</td>
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<tr>
<td>1100</td>
<td>0.60</td>
<td>5.2</td>
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</tr>
<tr>
<td></td>
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</tr>
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<td>2.8</td>
</tr>
<tr>
<td></td>
<td>4.75</td>
<td>3.1</td>
</tr>
</tbody>
</table>
Nickel temp. = 485°C

Figure 4.4 Mean void and loop radii vs dose
Nickel temp. = 485°C

Figure 4.5 Void and dislocation line density vs Dose
4.5 and tabulated in table 4.2. The magnitude of the error in the dislocation line density depends on the error in the estimate of the total foil thickness. This error has been minimised by counting the number of thickness fringes near the $s = 0$ position (87). According to Hirsch et. al. (87) foil thickness can be measured to an accuracy of $\pm$ 5% in this condition.

In a 1MeV microscope it is almost impossible to operate with only two beams as systematic reflections always occur strongly due to the flatness of the Ewald sphere. Thus there will be an error in the magnitude of the extinction distance used to calculate the foil thickness. The total error in the measurement of foil thickness has been estimated to be about 20% which is translated directly to be the error in the dislocation line density. This is in accordance with the estimate by Ham (86).

The estimate of void and dislocation loop densities depends on the accuracy obtained using stereomicroscopy. It has been pointed out by Hirsch et. al. that the $\pm$ 5% error claimed by Nankivell (101) can only be achieved with careful attention to the microscope tilt holder. By matching a large number of voids in the stereo pair it is felt that the error in estimating the defect layer width has been minimised but will still be of the order of 10%.

Measuring the void dimension by taking the radius of the smallest sphere which would fit over it would give a large error in calculating the extent of volume swelling. This experiment however requires only information on the increase in the void size and so this is a convenient parameter to measure. The void radii can be
measured to an accuracy of ± 0.05mm on the micrographs which are printed to 60K magnification. Thus the error in the void size measurements is of the order of 20Å.

Loop radii measurement is less accurate as the width of the dark band from which measurements were made is variable. Figure 4.2(a) shows some loops with no dark band along their longer axis and some with a dark band. Measurements of loop diameter were always made to the inside of this dark band, if it was visible. The loop diameter could be measured to an accuracy of ± 0.25mm on the plate which corresponds to ± 40Å on the specimen.

The data to be input to the program is summarised, for three temperatures, in table 4.3. The experiment carried out at 440°C was not continued into the void regime. Table 4.3 does not include the estimated errors as it shows the specific values which were used to simulate the microstructural development.

4.1.2. Analysis of output

Figure 4.6 shows a typical page of output from the program. The output is searched to find, for each value of vacancy migration energy, a calculated saturation time close to the observed saturation time. The interstitial relaxation volume and vacancy migration energies for which this condition holds are noted. These are tabulated for the three temperatures in table 4.4 and plotted as figure 4.7. The quoted values of $\Delta V_\text{I}$ are ± 0.025Å. Each curve in this figure represents pairs of values of $\Delta V_\text{I}$ and $E^V_m$ which fit the loop growth data at a particular temperature. The values of $\Delta V_\text{I}$ and $E^V_m$
<table>
<thead>
<tr>
<th></th>
<th>440</th>
<th>485</th>
<th>525</th>
</tr>
</thead>
<tbody>
<tr>
<td>Irradiation temperature / °C</td>
<td>440</td>
<td>485</td>
<td>525</td>
</tr>
<tr>
<td>Loop density / * 10 cm</td>
<td>2.9</td>
<td>1.76</td>
<td>0.40</td>
</tr>
<tr>
<td>Interaction radius / A</td>
<td>930</td>
<td>1100</td>
<td>1820</td>
</tr>
<tr>
<td>Saturation time / s</td>
<td>660</td>
<td>300</td>
<td>413</td>
</tr>
<tr>
<td>Growth rate / A s⁻¹</td>
<td>1.36</td>
<td>3.7</td>
<td>4.4</td>
</tr>
<tr>
<td>Max. dislocation density / *10⁵ cm cm⁻³</td>
<td>1.7</td>
<td>1.2</td>
<td>0.45</td>
</tr>
<tr>
<td>Min. dislocation density / *10⁵ cm cm⁻³</td>
<td>4.0</td>
<td>2.0</td>
<td>2.5</td>
</tr>
<tr>
<td>Foil thickness / A</td>
<td>4900</td>
<td>4300</td>
<td>5000</td>
</tr>
<tr>
<td>Dose rate / 10³ d.p.a.s⁻¹</td>
<td>2.0</td>
<td>2.0</td>
<td>2.0</td>
</tr>
<tr>
<td>Dose at which voids appear / d.p.a.</td>
<td>---</td>
<td>2.1</td>
<td>3.6</td>
</tr>
<tr>
<td>Radius with which voids appear / A</td>
<td>---</td>
<td>56</td>
<td>95</td>
</tr>
</tbody>
</table>

Table 4.3 Void and loop growth data
**Fig. 4.6** Typical page of output from the radiation damage simulation program.

<table>
<thead>
<tr>
<th>DOSE</th>
<th>VOID RADIUS</th>
<th>OBS VOID RADIUS</th>
<th>LOOP RADIUS</th>
<th>( F(\eta) )</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.62</td>
<td>0.0</td>
<td>0.0</td>
<td>1038.86</td>
<td>0.16</td>
</tr>
<tr>
<td>2.00</td>
<td>0.0</td>
<td>0.0</td>
<td>1038.86</td>
<td>0.16</td>
</tr>
<tr>
<td>3.22</td>
<td>50.00</td>
<td>50.00</td>
<td>1038.86</td>
<td>0.16</td>
</tr>
<tr>
<td>4.00</td>
<td>80.62</td>
<td>85.00</td>
<td>1038.86</td>
<td>0.33</td>
</tr>
<tr>
<td>5.00</td>
<td>109.90</td>
<td>97.00</td>
<td>1338.86</td>
<td>0.34</td>
</tr>
<tr>
<td>6.00</td>
<td>124.77</td>
<td>104.00</td>
<td>1038.86</td>
<td>0.37</td>
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<tr>
<td>7.00</td>
<td>137.07</td>
<td>106.00</td>
<td>1038.86</td>
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<tr>
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<td>147.47</td>
<td>110.00</td>
<td>1038.86</td>
<td>0.42</td>
</tr>
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</table>

Bias = 0.152634

Loop Sat. of the DOSE = 0.395440 @ 000000000000
Calc Sat. Time = 0.1975000 @ 030000000000
OBS Sat. Time = 0.3000000 @ 030000000000

ZI = 2.1481972E01
ZV = 0.125577E01
EM = 1.1999999
RI = 0.2420333E-07
TH = 0.440199E-04
VI = 0.355300
<table>
<thead>
<tr>
<th>Irradiation temperature/°C</th>
<th>440</th>
<th>485</th>
<th>525</th>
</tr>
</thead>
<tbody>
<tr>
<td>$E_n^\gamma$/eV</td>
<td>$\Delta V_I/\Omega$</td>
<td>$\Delta V_I/\Omega$</td>
<td>$\Delta V_I/\Omega$</td>
</tr>
<tr>
<td>1.00</td>
<td>0.225</td>
<td>0.225</td>
<td>0.225</td>
</tr>
<tr>
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<td>0.225</td>
<td>0.225</td>
</tr>
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<td>0.225</td>
<td>0.25</td>
<td>0.25</td>
</tr>
<tr>
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<td>0.225</td>
<td>0.275</td>
<td>0.25</td>
</tr>
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<td>0.225</td>
<td>0.30</td>
<td>0.25</td>
</tr>
<tr>
<td>1.25</td>
<td>0.25</td>
<td>0.325</td>
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</tr>
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<td>1.30</td>
<td>0.25</td>
<td>0.35</td>
<td>0.30</td>
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<tr>
<td>1.35</td>
<td>0.275</td>
<td>0.425</td>
<td>0.35</td>
</tr>
<tr>
<td>1.40</td>
<td>0.30</td>
<td>0.50</td>
<td>0.40</td>
</tr>
<tr>
<td>1.45</td>
<td>0.35</td>
<td>0.60</td>
<td>0.55</td>
</tr>
<tr>
<td>1.50</td>
<td>0.475</td>
<td>0.80</td>
<td>0.75</td>
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<td>0.65</td>
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<td>—</td>
</tr>
<tr>
<td>1.60</td>
<td>0.825</td>
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</table>

Table 4.4 Values of Interstitial relaxation Volume and Vacancy migration energy.
Figure 4.7 $\Delta V x$ vs $E'_m$
where the curves cross satisfy the loop growth rate at all three temperatures. Thus a value for the bias, $B_0$, and $E_{\text{m}}$ can be found irrespective of the precise variation of $B$ and $E_{\text{m}}$ with temperature. From figure 4.7 it can be seen that the curves cross over with $E_{\text{m}}$ between 1.1eV and 1.2eV which corresponds to $E_0$ to $E_1$ in the figure. The corresponding value of $\Delta V_\text{F}$ is about 0.25 ($\pm$ 0.025)$A$ i.e. $F$ to $F_1$ in the figure. Thus $E_{\text{m}}$ = 1.15 ($\pm$ 0.05)eV and $\Delta V_\text{F}$ = 0.25$A$. The bias corresponding to these values is 6.5%.

The value of $E_{\text{m}}$ is in good agreement with that of 1.2($\pm$ 0.2)eV found by Dlubek et. al. (102) using a positron annihilation method. It also agrees with the values of $\equiv$1.2eV given by Yoo and Stiegler (90) and Kiritani and Takata (7). Yoo and Stiegler found the interstitial bias to be 6% for nickel which again agrees well with the value obtained in this work.

Having determined $E_{\text{m}}$ and $\Delta V_\text{F}$ from the loop growth data the subsequent void growth may be analysed in detail. This has been carried out for the irradiations at 485°C and 525°C. Figure 4.8 and table 4.5 show the experimental results at the two temperatures and the computed results assuming a simple diffusion controlled mechanism for void growth. The agreement between the computed and experimental curves is reasonable, especially at low doses. At higher doses however the computed curve rises above the experimental one. Fisher et. al. found their calculated curve to be higher than the experimental in their analysis of void growth in copper. They suggested increasing the value of the interface radius ($r^*$ in equation 1.15) to lower the calculated growth rate. This implies that the growth is being modelled as surface controlled diffusion.
<table>
<thead>
<tr>
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<td></td>
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<td>exp.</td>
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<td>—</td>
<td>108</td>
<td>7.0</td>
</tr>
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<td>7.0</td>
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<td>114</td>
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<tr>
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<td>—</td>
</tr>
<tr>
<td>8.0</td>
<td>—</td>
<td>119</td>
<td>—</td>
</tr>
</tbody>
</table>

Table 4.5  Observed and calculated void radii at two irradiation temperatures
Simple diffusion control at 485°C and 525°C

- △ 485°C exp.
- ▽ 485°C theory
- □ 525°C exp.
- ◊ 525°C theory

Error bar

$\Delta V_x = 0.25 \Omega$

$E_m^v = 1.15 \text{eV}$

Dose /d.p.a.

Figure 4.8 Void growth curves
Table 4.6 Void growth data for varying interface radii.

<table>
<thead>
<tr>
<th>Dose /d.p.a.</th>
<th>Void radii / A</th>
<th>Interface radii / A</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Exp.</td>
<td>2.5</td>
</tr>
<tr>
<td>2.1</td>
<td>56</td>
<td>—</td>
</tr>
<tr>
<td>3</td>
<td>—</td>
<td>67</td>
</tr>
<tr>
<td>3.6</td>
<td>74</td>
<td>—</td>
</tr>
<tr>
<td>4</td>
<td>—</td>
<td>85</td>
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<tr>
<td>4.1</td>
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</tr>
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<td>4.7</td>
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<td>108</td>
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<tr>
<td>6.3</td>
<td>105</td>
<td>—</td>
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<tr>
<td>7</td>
<td>—</td>
<td>117</td>
</tr>
<tr>
<td>7.2</td>
<td>110</td>
<td>—</td>
</tr>
<tr>
<td>8</td>
<td>—</td>
<td>124</td>
</tr>
</tbody>
</table>
Varying interface radii  $485^\circ C$

$\Delta V_I = 0.25 \Omega$

$E^V = 1.15eV$

Figure 4.9 Void growth curves
Figure 4.9 and table 4.6 show the experimental data points at 485° C along with those computed with various values of $r^*$. The agreement between the calculated and experimental growth curves is best with $r^* = 100A$. This is of the same order as found by Fisher et. al. [9].

Assuming an interface controlled mechanism may not be the best explanation. This factor enters the equation as:

$$ Y = \frac{4\pi r_v^2}{r_v + r^*} $$

Thus as the void radius increases the $r_v^2$ term becomes dominant. This means that the model will not change the calculated growth rate in such a way as to lower the growth rate at large radii more than at small radii.

It should also be noted that this model involves a thin spherical shell surrounding a void which modifies the capture efficiency of the void for point defects. As large voids are strongly faceted this model may not be applicable.

An alternative approach would be to assert that voids may, as do dislocations, have a preference for interstitials over vacancies. This preference for interstitials could, as with dislocations, be due to the stronger interaction with an interstitial than a vacancy due to either size effect interactions or stress field interactions. The void bias is likely to be less than the dislocation bias and thus more vacancies than interstitials would still arrive at void surfaces because, although the interstitials are more strongly attracted to the void than vacancies, there would be fewer interstitials available as they are even more strongly attracted to the dislocations. This
possibility has been investigated with the results shown in figure 4.10 and tabulated in table 4.7. Agreement between the experimental and calculated growth curve is very good under this void bias condition. The void bias used was 0.5% for interstitials. Mansur and Wolfer (103) investigated the effect of solute segregation to voids by calculating the change in the capture efficiencies for point defects of voids with, and without, a surrounding shell. During the course of this work they suggest that a void with no surrounding shell has a large preference for interstitials over vacancies. For voids of radius greater than 40Å they show a value of the order of 5% to 10% for this bias.

It was stated in section 3.1 that measurement of defect bias and vacancy diffusivity are the major problems set in equations 1.12 to 1.18. In nickel this is not strictly true as although there is some controversy over the value of the bias the value of the vacancy migration energy is well established. This gives a check on the validity of the technique of Fisher et. al. The success of the method in arriving at both the accepted value of $E_v$ and also at the likely value of the bias means that the technique may be applied to other materials in which the parameters are not so well known.

Bullough and Nelson (79) conclude that the elastic interaction between point defects and voids is unlikely to cause a significant drift flow of point defects to voids. They base this calculation on the fact that as a point defect approaches a void the void surface can eventually be modelled as a plane free surface. This may be true for point defects approaching the void faces but for those approaching the edges and corners, where the deformation due to
<table>
<thead>
<tr>
<th>Dose /d.p.a.</th>
<th>Void radii / Å</th>
<th>experimental</th>
<th>calculated with void bias = 0.5%</th>
</tr>
</thead>
<tbody>
<tr>
<td>2.1</td>
<td>56</td>
<td>—</td>
<td>—</td>
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<tr>
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<tr>
<td>4</td>
<td>86</td>
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<td>—</td>
</tr>
<tr>
<td>4.7</td>
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</tr>
<tr>
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</tr>
<tr>
<td>6</td>
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</tr>
<tr>
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</tr>
<tr>
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<td>—</td>
<td>110</td>
<td>—</td>
</tr>
<tr>
<td>7.2</td>
<td>110</td>
<td>—</td>
<td>—</td>
</tr>
<tr>
<td>8</td>
<td>—</td>
<td>115</td>
<td>—</td>
</tr>
</tbody>
</table>

Table 4.7 Void growth data calculated with a void bias of 0.5%
Simple diffusion control Void bias = 0.5%

- ΔV_i = 0.25\,\Omega
- E^V = 1.15\,\text{eV}

- experimental
- Void bias of 0.5% for interstitials over vacancies.

Dose /d.p.a.

Figure 4.10 Void growth curves 485°C
strain may be higher, this may not be the case. More detailed analysis of the form of the strain field surrounding large faceted voids is necessary before conclusions can be drawn, with any confidence, regarding the void to point defect interactions.
CHAPTER FIVE

COMPUTATIONAL METHODS
Three contrast mechanisms contribute to the image seen in an electron microscope. These are thickness contrast, diffraction contrast due to the deformation of the host lattice around the defect and defocusing contrast. In the literature there are no simulations of image contrast which consider all three of these mechanisms for realistic morphologies.

In order to rectify this situation the multislice matrix method of Thölen \[65\] has been adapted assuming both the column approximation and the two beam approximation to be valid. Computations have been carried out for three strain conditions:

(a) Brick or cube shaped cavities with no surrounding strain field

(b) Small spherical or cubic cavities surrounded by a spherically symmetric strain field

(c) Brick or cube shaped cavities which have a non-spherically symmetric strain field associated with them.

The parameters which must be variable in an image simulation program are listed below:

(1) Defect size and shape

(2) Defect depth

(3) Diffraction vector operating \(\mathbf{g}\)

(4) Deviation from the Bragg condition \(\mathbf{g}\)

(5) Foil thickness and orientation

(6) Extinction distance

(7) Focussing conditions and microscope aberrations.
These parameters will be discussed in the context of case (a) but they will clearly appear in the other two cases.

5.1. Cavities with no associated strain field

The technique used does not exclude spherical cavities but as they are not the most interesting shape only a small amount of effort has been spent on them. The adaptation of Thölén's method is represented by figure 5.1. As there is no strain field present the absorption and scattering matrices $A_x$ need only be calculated once as the effective $\sigma$ value will remain unchanged through the foil. Taking column 1 as an example the wave matrix at the bottom of the first slice is $A_x$, which then becomes the incident wave at the top of the second slice. The wave matrix at the bottom of the second slice is $A_x A_x$. As the absorption matrix is the same for this slice. For column 1 this process is repeated until the lower foil surface is reached.

Column 2 however travels through the cavity and so must be treated differently. Again the absorption and scattering matrix is the same as that for column 1, say $A$. Thus the wave incident at the top of the second slice is $A_x A_x$ and that at the bottom of this slice is $A_x A_x A_x$. This procedure is repeated until the cavity is encountered. It is then assumed that only the phase of the wave changes until the matrix material is again encountered. For example if the wave matrix at a point $Q$ on the diagram is $\phi$, then the wave matrix at $Q'$ is also taken to be $\phi$. This step incorporates the difference in the amount of normal absorption suffered by an electron beam passing through the defect to one passing only through perfect crystal. Thus the multiplication suggested in equation 2.7 is unnecessary in this
Figure 5.1 Multislice model
formulation. The procedure of successive multiplication by the 'A' matrices then begins again until the lower foil surface is reached. If the in focus image is required the phase change occurring between \( Q \) and \( Q' \) is irrelevant as only the intensity is needed. The intensity is the wave multiplied by its complex conjugate so the phase term disappears.

It is clear from the preceding discussion that there must be some means of deciding if the point being considered is inside or outside the cavity. Defect size, shape and depth are variable in the test for this condition. When the centre of the cavity is coincident with the foil origin the coordinates of the point in terms of the foil axes can be transformed to points in terms of the cavity axes by:

\[
[h \ k \ l]_v = T_{ij} \begin{pmatrix} h \\ k \\ l \end{pmatrix}_f
\]

\[
T_{ij} = \frac{1}{AB} \begin{pmatrix} 0 & k + l & hB \\ -A1 & -h & kB \\ kA & -h & lB \end{pmatrix}
\]

where

\[
A = (h^2 + k^2 + l^2)^{\frac{1}{2}}
\]

\[
B = (l^2 + k^2)^{\frac{1}{2}}
\]

\([h \ k \ l]_v \) represent the coordinates in terms of the void axes

\([h \ k \ l]_f \) represent the coordinates in terms of the foil axes.

If the cavity centre is not positioned in the middle of the foil but at a depth, \( D_e \), the transformation must be altered. This situation is represented by figure 5.2. The coordinates of the cavity centre in foil terms are \((0,0,-D_m)\) where \( D_m \) is given by:

\[
D_m = \frac{t}{2} - D_e.
\]

Therefore:
Figure 5.2 Axis transformation
A special case is when \([h k l] = [N 0 0]\). In this case, if the void origin is at the foil centre, the void axes are coincident with the foil axes and so no transformation is required. If the origins are not coincident but the void is at depth \(D_e\) only the z void coordinate must be changed as in the general case.

Defect size and shape are variable in the test to decide if the point in question is inside or outside the cavity. If the coordinates of a point in terms of the void axes are \(X'_v, Y'_v, Z'_v\) the test to decide if the point is inside a spherical void of radius \(R\) would be:

\[
\sqrt{(|X'_v|^2 + |Y'_v|^2 + |Z'_v|^2)^{1/4}} \leq R
\]

A similar test is carried out for cubic or brick shaped cavities where the defect dimensions are input into the program.

In the case where no strain field is present the operating \(\mathbf{g}\) vector merely enters the program via the value of the extinction distance to be input. The value of the deviation from the Bragg condition is input into the program in terms of \(w = s\frac{\xi}{J_3}\). Foil thickness and orientation are important parameters which are input with the foil orientation being the \([h k l]\) in the transformation matrix. The value of the extinction distance at \(s = 0\) is input to the program and changed to the effective extinction distance via equation 2.16.
The focusing conditions and lens aberrations are incorporated in a similar manner to that used by Rühle and Wilkens [67]. A detailed account of transfer theory was given by Hanszen [94] and references 95 and 96 are introductory texts for Fourier theory.

5.2. Defocussing effects and lens aberrations

The principles of this method were discussed in section 2.1 and represented in figure 2.4. For weak phase objects and in the isoplanatic approximation the influence of defocussing can be calculated in three steps.

The Fourier transform of the amplitude existing at the lower foil surface, say \( z = t' \) is calculated by:

\[
\tilde{\psi}(t',\mathbf{p}) = \int \psi(t',\mathbf{r})\exp(2\pi i \mathbf{p} \cdot \mathbf{r}) \, d\mathbf{r}
\]

Where \( \mathbf{p} \) is a two dimensional vector parallel to the x-y plane in Fourier space and \( \mathbf{r} \) is a position vector in the x-y plane in real space.

The amplitudes of the Fourier partial waves have to be multiplied by a phase factor \( T(\mathbf{p}) \). This phase factor is known as the amplitude transfer function and accounts for the phase shifts of the Fourier partial waves propagating from the plane \( z = t/2 \) to \( z = t/2 + \Delta f \).

\[
T(\mathbf{p}) = \exp(2\pi i W(\mathbf{p})/\lambda) \cdot B(\mathbf{p})
\]

where \( B(\mathbf{p}) \) is the aperture function and \( W(\mathbf{p}) \) is the wave aberration function. The function \( W(\mathbf{p}) \) can include the effects due to spherical aberration, axial astigmatism and the defocussing effect.

\[
W(\mathbf{p}) = C_s \frac{\mathbf{p}^2}{4} - \frac{\Delta f}{2} \mathbf{p}_x^2 - C_a (\mathbf{p}_x^2 - \mathbf{p}_y^2)
\]
C is the axial astigmatism

\( p_x, p_y \) are the components of \( \mathbf{p} \) parallel to the \( x \) and \( y \) axis respectively.

\[ B(p) = 1 \text{ for } p \leq p_{\text{max}} \]

\[ B(p) = 0 \text{ for } p > p_{\text{max}} \]

where \( p_{\text{max}} \) is the semi-angle of the objective aperture.

The influence of partial spatial coherence, due to the finite size of the electron source, and partial temporal coherence can be inserted into the program at this stage. Partial temporal coherence (chromatic aberration) is due to the electrons leaving the gun with a Maxwellian distribution of energy. The effects of spatial coherence on the coherent transfer function may be approximated by an envelope function which does not effect the phase of the coherent transfer function \( [97] \). This envelope function is given, in the absence of astigmatism, by:

\[ E_S(p) = \exp\left(-\frac{1}{2}q_0(C_{cp}p_\lambda^3 - \Delta f p_\lambda^2)\right) \quad \text{5.7} \]

for a Gaussian source where \( q_0 \) is the effective source half width and the other symbols are as previously defined.

The effects of the finite energy spread of the beam may also be represented by an envelope function. This function is given by Hanszen and Trepte \( [98] \) as:

\[ E_t(p) = \exp\left(-\frac{1}{2}\left(C_{\Delta E} p_\lambda^3\right)/16\ln(2)E^2\right) \quad \text{5.8} \]

where \( C_{\Delta E} \) is the chromatic aberration coefficient

\( \Delta E \) is the full width at half height of the electron energy distribution

and \( E \) is the mean electron energy.
The total transfer function is given by:

\[ T(p) = T'(p).E_g(p).E_t(p) \]  \hspace{1cm} 5.9

To calculate the amplitude field \( \psi(t/2+\Delta f, \tau) \) at the plane \( z = (t/2+\Delta f) \) an inverse Fourier transform must be performed.

\[ \psi(t/2+\Delta f, \tau) = \int \tilde{\psi}(t/2, p).T(p)\exp(-2\pi ip.\tau)dp \]  \hspace{1cm} 5.10

Rühlle and Wilkens (67) imposed a condition of rotational symmetry on the cavity such that an analytical solution could be found for this function. However this is an unnecessary restriction as using fast Fourier transform routines the integrations can be performed numerically, see also (99). Thus cavities of various shapes can be conveniently treated. As Fourier transforms operate in the complex domain and the transfer function is complex the phase of the wave at the lower foil surface becomes important in these calculations. Thus allowance must be made for the phase change of the beam in passing through the cavity.

5.3. Spherical strain field

When including the effect of strain fields in contrast calculations the method closely follows that of Thölén (65) discussed in section 2.1. The strain field enters the calculation in the determination of the effective deviation parameter given by:

\[ w_{e_t} = w + \frac{\partial}{\partial \alpha} \beta' \]

From equation 2.2

\[ \beta' = g.d[R(z)] \frac{dz}{dz} \]

where \( R(z) \) is the displacement function due to the strain field of
the defect. The model is represented by figure 5.3. The displacement field used was that used by Brown and Mazey (100) for inert gas bubbles in solids. The displacement, $R$, is given by:

$$ R = \frac{\epsilon \gamma^3}{\rho^2} $$  \hspace{1cm} \text{(5.11)}

with

$$ \epsilon = \frac{(-2\gamma + Pr_c)}{4\sqrt{\gamma}} $$  \hspace{1cm} \text{(5.12)}

where $\gamma$ is the shear modulus,

$\gamma$ is the surface energy,

$P$ is the gas pressure in the bubble,

$r_c$ is the bubble radius,

and $\rho$ is the distance of the point under consideration from the centre of the bubble.

In column 1 of the diagram the displacement is calculated to be $R_1$ at the top of the first slice. At the bottom of this slice the displacement is again calculated and is found to be $R_2$. In order to find $g.dR(z)/dz$ we then take:

$$ \frac{g.dR(z)}{dz} = g.(R_2 - R_1) \Delta z $$  \hspace{1cm} \text{(5.13)}

This value is used to calculate the $\mathbf{w}_{\text{eff}}$ at the top of the first slice which is put into equation 2.11 to calculate the scattering matrix $A_1$ in the first slice. The process is then repeated until the lower foil surface is reached. Column 2 passes through the cavity and the approach is similar to the 'no strain' case. The $A_1$ matrices are calculated until the column reaches the cavity and then the beam amplitude is assumed to be unchanged until it leaves the cavity. The process of calculating the $A_1$ is then resumed until the bottom of the foil is reached.
Figure 5.3 Spherical strain field model
When considering the strain fields surrounding defects the operating vector $a$ is input to the program and used in the calculation of equation 5.13.

5.4. Non-spherical strain fields

The technique used for non-spherical strain fields follows exactly that used for spherical strain fields apart from in the calculation of $R(z)$, the displacement function.

The model chosen to represent the cavity and forces is shown in figure 5.4. The forces exerted by the gas in the bubble were represented as sheets of force acting on each face of the cube, or brick shaped bubbles. The displacement at each point in the foil was calculated using the Green's function method discussed in section 2.2. Using the perfect lattice Green's function i.e. ignoring the missing material within the cavity will result in an overestimate of the surrounding strain field(111). Tewary and Bullough[112] give a defect Green's function formulation which allows for this missing material. The image simulation program has been developed to allow insertion of both this factor and anisotropy of the matrix material. Due to the constraint of realistic computation time neither of these conditions have been included in the present formulation. The displacement at a general point $r$ in direction $i$, say $U_i$, due to a force centred at $r'$ in direction $j$, say $F_j$, is given by:

$$U_i (r) = G_{ij}(r-r')F_j(r')$$  \hspace{1cm} 5.14

For an isotropic body the Green's function $G_{ij}(r)$ can be expressed in the form (79):
Fig. 5.4 Model of a "brick shaped" pore showing the pore axes and face names.
\[ G_{ij}(r) = \frac{1}{16\pi(1-v)} \left[ 2(1-v)\delta_{ij} \left| r_{pp} - r_{ij} \right| \right] \quad 5.15 \]

In equation 5.15 the comma notation denotes partial differentiation, i.e. \( r_{pp} = \nabla r \) and \( \delta_{ij} \) is the Kronecker Delta function. From 5.15 it is clear that

\[ G_{ij}(R) = \frac{1}{16\pi(1-v)} \left[ 2(1-v)\delta_{ij} R_{pp} - R_{ij} \right] \quad 5.16 \]

where \( R = r - r' \) and \( R = |R| \).

Say \( r = (x_1, x_2, x_3) \) and \( r' = (x'_1, x'_2, x'_3) \), then:

\[ R = \left[ (x_1-x'_1)^2 + (x_2-x'_2)^2 + (x_3-x'_3)^2 \right]^{\frac{1}{2}} \quad 5.17 \]

\[ R_{pp} = \frac{2}{R} \quad \text{and} \quad R_{ij} = \frac{\delta_{ij} - x_i x_j}{R^3} \]

Therefore \[ G_{ij}(R) = \frac{1}{16\pi(1-v)} \left[ 2(1-v)\delta_{ij} + \frac{x_i x_j}{R^3} \right] \quad 5.18 \]

Three methods have been used to calculate the lattice displacement using the Green's function formulation. The first of these methods was merely to consider a set of point forces acting outwards at the origin along the \(<100>\). This method is computationally simple but was used only as a test of the Green's function calculation routine as it is not a good physical model of a general pore. It is only a reasonable model if the pore in question is extremely small such that the forces on each face are effectively acting along the axes. The two methods which were concentrated on were a finite element technique discussed in section 5.4.1 and an analytical approach discussed in section 5.4.2.
5.4.1 Finite element method

This technique involves modelling each element of the pore faces as having a force acting outwards from the centre of the element parallel to the face normal. The model is represented as figure 5.5. In figure 5.5(a) the face on the ZY plane is shown. The dots in the diagram represent the forces acting 'out of the paper' parallel to the X-axis which is marked by the square. This diagram represents all of the pore faces. In the figure (b) is a projection of (a) which may make the model more transparent. The origins of the forces on each element must be calculated within the program as they depend on the how many elements the hole is in size. Four possible situations are shown as figure 5.5(c).

If there is an odd number of steps in direction 'a' then the 'a' coordinate of the origins of the forces will be at $\pm na$, where $n$ is an integer. If there is an even number of steplengths then the 'a' coordinate will be given by $\pm(n+0.5)$, where $n$ is an integer. This unfortunately means that the program can only deal with pores which are an integral number of steplengths in each direction. However this is not a large restriction as the program has been written to allow any hole size to be input and the dimensions will then be altered to the nearest integral multiple of the steplengths in each direction.

This method is mathematically relatively simple as it only involves calculating the $G_{ij}(x-x')$ at each point within the lattice using the origin of each force as $x'$ in turn. Computationally this is again simple as it just implies repeated passes through the calculation routines with the parameters changed. It must be stressed
Fig. 5.5 Force origins on pore face
however that for a large pore, say 200*200*200A in size the procedure will be very time consuming. If the steplength is about 20A in each direction each face will have 100 elements on it and thus 100 origins of force. Performing the calculation with the minimum lattice size, say 64*64*50 points with this hole size will involve about $10^9$ passes through the routine. These computations have only been carried out in full on the FPS120B array processor at Surrey University. Timing and accuracy checks have been carried out on the Prime 750's at Surrey[113] and also on the VAX 11/780 at Queen Elizabeth College. These checks have also been carried out for the analytical approach which will now be discussed.

5.4.2 Analytical approach

In this method the displacement at each lattice point is calculated analytically by first calculating that due to a general point on a face. This point is then allowed to vary over the two dimensions of the face by double integration. The procedure is repeated for each face in turn. Mathematically and physically this is a more elegant technique but unfortunately it is a very complex procedure computationally. The details of the calculation are given below.

Referring to figure 5.4 the coordinates of general points on each face of the hole are:

on face $A^+$ $r' = a, x'_1, x'_3$

$A^- r' = -a, x'_1, x'_3$

$B^+ r' = x'_1, b, x'_3$

$B^- r' = x'_1, -b, x'_3$

$C^+ r' = x'_1, x'_2, c$
Let \( |F| = P^* \) be the pressure of gas within the hole multiplied by the area of the face on which this pressure is acting and represent the factor \( 1/16\pi(1 - \nu) \) by \( L \).

Then the displacements due to the stress on faces \( A^+ \) and \( A^- \) are given by:

\[
U^+_{\iota} = P^* L \int_{R^+} \left( \frac{[3 - 4 \nu] \delta_{ij}}{R^+} + \frac{X^+_{\iota} (x_i - a)}{(R^+)^2} \right) dx_1^i \ dx_3^j \\
U^-_{\iota} = -P^* L \int_{R^-} \left( \frac{[3 - 4 \nu] \delta_{ij}}{R^-} + \frac{X^-_{\iota} (x_i + a)}{(R^-)^2} \right) dx_1^i \ dx_3^j
\]  

5.19
5.20

where \( U_{\iota} \) is the displacement in direction \( \iota \)

\[ R^+ = \left( (x_i - a)^2 + (x_2 - x_2^i)^2 + (x_3 - x_3^i)^2 \right)^{\frac{1}{2}} \]

and \( X^+_{\iota} = (x_i - a), (x_2 - x_2^i), (x_3 - x_3^i) \).

Clearly equations 5.19 and 5.20 imply that the displacement in direction \( \iota \), due to the stress on face \( A^+ \), is obtained by calculating the Green's function times the force acting at a general point on the \( A^+ \) face and then integrating this over the area of the face. The other components of the displacement due to the stresses on faces \( B^+, B^- \) and \( C^+, C^- \) are obtained using 5.19 and 5.20. The required displacement field is given by:

\[
U_{\iota} (x_1^i, x_2^i, x_3^i) = P^* L \left[ \left[ I_{\iota} (a) - I_{\iota} (-a) \right] + \left[ I_{\iota} (b) - I_{\iota} (-b) \right] \right. \\
\left. + \left[ I_{\iota} (c) - I_{\iota} (-c) \right] \right]  \\
\]

5.21

where \( I_{\iota} (a) = \int_{-b}^{+b} \int_{-c}^{+c} \left( \frac{[3 - 4 \nu] \delta_{ij}}{R_i} + \frac{X_{\iota} (x_i - a)}{R_3} \right) dx_1^i \ dx_3^j \)

with \( R_1 = \left( (x_i - a)^2 + (x_2 - x_2^i)^2 + (x_3 - x_3^i)^2 \right)^{\frac{1}{2}} \)

and \( X_{\iota} = (x_i - a), (x_2 - x_2^i), (x_3 - x_3^i) \).
\[
I_i^2(b) = \int_{-a}^{+a} \int_{-c}^{+c} \frac{(3-4\nu)\delta_{ij} + X_{ij}(x_j - b)}{R_i^2} \, dx_j \, dx_i
\]
with
\[
R_i = \sqrt{(x_i - x_i')^2 + (x_j - b)^2 + (x_k - c)^2} \]
and
\[
X_{ij} = \{ [x_i - x_i']^2 + (x_j - b)^2 + (x_k - c)^2 \}^{\frac{1}{2}}
\]

Using the fact that \( \frac{\partial}{\partial x_i}(\frac{1}{R_i}) = -\frac{X_i}{R_i^3} \) the integrals may be rewritten:
\[
I_i^1(a) = \int_{-b}^{+b} \int_{-c}^{+c} \frac{(3-4\nu)\delta_{ij} - (x_i - a)}{R_i^2} \, dx_j \, dx_i
\]
\[
I_i^2(b) = \int_{-a}^{+a} \int_{-c}^{+c} \frac{(3-4\nu)\delta_{ij} - (x_j - b)}{R_i^2} \, dx_j \, dx_i
\]
\[
I_i^3(c) = \int_{-a}^{+a} \int_{-b}^{+b} \frac{(3-4\nu)\delta_{ij} - (x_k - c)}{R_i^2} \, dx_j \, dx_i
\]

These may in turn be expressed as:
\[
I_i^1(a) = \int_{-b}^{+b} \int_{-c}^{+c} \frac{(3-4\nu)\delta_{ij} - (x_i - a)}{R_i^2} \, dx_j \, dx_i
\]
\[
I_i^2(b) = \int_{-a}^{+a} \int_{-c}^{+c} \frac{(3-4\nu)\delta_{ij} - (x_j - b)}{R_i^2} \, dx_j \, dx_i
\]
\[
I_i^3(c) = \int_{-a}^{+a} \int_{-b}^{+b} \frac{(3-4\nu)\delta_{ij} - (x_k - c)}{R_i^2} \, dx_j \, dx_i
\]

It is then clear that:
\[
J^2 = J^1(x_i, x_j, abc)
\]
\[
J^3 = J^1(abc)
\]

Thus the only integral required is \( J^1(x_i, x_j, abc) \) which has the form:
\[
J^1(x_i, x_j, abc) = \int_{-a}^{+a} \int_{-c}^{+c} \frac{dx_i}{[(x_i - a)^2 + (x_j - x_j')^2 + (x_k - c)^2]^{\frac{1}{2}}}
\]

This integration is very lengthy but can be performed analytically with the result that equation 5.21 can be used to calculate the displacement field. The integration is given as appendix 1. Initially it has been assumed that the forces on the hole
faces are the product of an excess gas pressure times the area of the element in question. While this model should be a reasonable approximation it must be pointed out that there are limitations. The discussion of the problems and inaccuracies of the methods used is in chapter 6 of the report.
CHAPTER SIX

IMAGE SIMULATION RESULTS AND DISCUSSION
6.1. Introduction

In this chapter results are presented from an image simulation program which allows inclusion of a strain field around the defect. Some of these results are compared to experimentally obtained images. Computed micrographs shown in this section were obtained by using either the microfilm plotting package, Picpac, at the university of London computing centre or the A.E.D. 256 grey level graphics device at Queen Elizabeth college. On the A.E.D. terminal there is about one pixel jitter on the display screen such that text on prints from this device look slightly out of focus. Graphs were plotted using the Dimfilm plotting routines and a Calcomp 81 plotter or direct onto microfilm using a combination of Dimfilm and Picpac. Figure 6.1 is a flow chart of the image simulation program in the equilibrium pressure case. A typical set of input data is given as table 6.1. The column approximation was used in all the simulations presented in this report. This approximation implies ignoring refraction occurring at both surfaces of the cavity which is in common with all other simulations in this field. In section 6.2.1 all simulated images assume an equilibrium pore while section 6.2.2 does not make this assumption.

6.2.1. Equilibrium bubble

The effects of varying the parameters in image simulation will be shown in this section for a pore containing gas at the equilibrium pressure.
START

Read details of foil, cavity, diffraction conditions, and defocussing

Calculate
\[ A = \begin{pmatrix} a_{11} & a_{12} \\ a_{21} & a_{22} \end{pmatrix} \]

Read in the wave matrix at the top foil surface \( \varnothing \)

Consider the first point in the foil

Convert from foil to void coordinates

Is the point inside the hole?

Has the foil bottom been reached?

Calculate and print the intensity at the lower foil surface, normalising to the background intensity

Take the Fourier transform at the lower foil surface, \( \tilde{F} \)

Calculate the transfer function at each point, \( T \)

Calculate \( \tilde{F} \cdot T \)

Take the inverse Fourier transform

Print the intensity at the defocussed plane

STOP

Fig. 6.1 Flowchart of simulation program.
<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Foil thickness/A</td>
<td>1140</td>
</tr>
<tr>
<td>Diffraction vector</td>
<td>1 1 3</td>
</tr>
<tr>
<td>Gas pressure/Nm²</td>
<td>0.80E+10</td>
</tr>
<tr>
<td>Surface energy/ Jm²</td>
<td>1.5</td>
</tr>
<tr>
<td>Surface stress/Nm²</td>
<td>3.75E+10</td>
</tr>
<tr>
<td>Foil orientation</td>
<td>1 1 1</td>
</tr>
<tr>
<td>Number of steps in z direction</td>
<td>60</td>
</tr>
<tr>
<td>Number of steps in x,y directions</td>
<td>64</td>
</tr>
<tr>
<td>Pore dimensions/A</td>
<td>200 200 200</td>
</tr>
<tr>
<td>Spherical aberration coefficient/m</td>
<td>1.8E-03</td>
</tr>
<tr>
<td>Electron energy/eV</td>
<td>1.0E+05</td>
</tr>
<tr>
<td>Inner potential/V</td>
<td>20</td>
</tr>
<tr>
<td>Depth of pore centre/A</td>
<td>570</td>
</tr>
<tr>
<td>Magnification factor</td>
<td>3</td>
</tr>
<tr>
<td>Wavelength of electrons/m</td>
<td>3.7E-12</td>
</tr>
<tr>
<td>Chromatic aberration coefficient/m</td>
<td>1.8E-03</td>
</tr>
<tr>
<td>Axial astigmatism constant/m</td>
<td>0</td>
</tr>
<tr>
<td>Extinction distance/A</td>
<td>261</td>
</tr>
<tr>
<td>Effective source half width/A</td>
<td>50</td>
</tr>
<tr>
<td>Deviation from the Bragg position</td>
<td>1.0</td>
</tr>
<tr>
<td>Defocussing distance/A</td>
<td>6000</td>
</tr>
<tr>
<td>Full width at half height of electron energy distribution/eV</td>
<td>2</td>
</tr>
</tbody>
</table>

Table 6.1 Typical set of input data to simulation program
Figure 6.2 shows a series of images at various values of defocus using a cubic pore of side 250Å and a <111> foil orientation. The bubble is situated in the centre of a foil of 4Å in thickness and the value of the deviation parameter is \( w = 1.0 \). An experimental through focal series of helium bubbles in vanadium is presented as figure 6.3. The magnification is 450K on the plate and the defocussing distance is about 0.6mm. Figure 6.3(1) is a plot of the data obtained from a stereo pair of the region including the area shown as figure 6.3(a). The technique used to analyse stereo images and the consequent errors are given in section 3.1.1. Equation 3.7 was used to obtain the bubble depths and the foil thickness could also be measured in this fashion using dirt particles on the surfaces. The relevant data is given below.

\[ \Theta = \text{angle between the pair of micrographs} = 5° \]

\[ M = \text{the magnification on both micrographs} = 70K \]

\[ \Delta Y = \text{the parallax between the foil top and bottom} = 5mm \]

Thus:

\[ \Delta h = \text{vertical separation between the foil top and} \]

\[ \text{foil bottom} = \frac{5 \times 10^{-7}}{2 \times 2.7 \times 10^3 \times \sin 2°30'} = 2120Å \]

Table 6.2 is a summary of the results obtained from analysis of the marked bubbles contained in figure 6.3. In order to convert the units of length to extinction distance units the appropriate extinction distance for iron has been used. This is because values of the extinction distances are not available for vanadium and iron has a similar crystal structure and atomic weight. All the numbers quoted in extinction distance units must be treated with some circumspection as the value of \( w \) is not known such that the effective extinction distance may vary from that quoted. Table 6.2 and figure 6.3 are used
Fig. 6.2  Simulated through focal series of a cubic pore in a \(<11\bar{1}\>) orientation.
This vanadium specimen was annealed for 8 hours at 1150°C.

Fig. 6.3 Through focal series of helium bubbles in vanadium.
<table>
<thead>
<tr>
<th>Bubble</th>
<th>Dimensions /mm.</th>
<th>Dimensions /A</th>
<th>Centre /mm.</th>
<th>Depth /A</th>
<th>Depth /$\xi$</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>10,5,8</td>
<td>210,105,168</td>
<td>249</td>
<td>424</td>
<td>1.57</td>
</tr>
<tr>
<td>B</td>
<td>9,10,8.5</td>
<td>189,210,178</td>
<td>248</td>
<td>828</td>
<td>3.14</td>
</tr>
<tr>
<td>C</td>
<td>7,3,5</td>
<td>147,63,105</td>
<td>247.75</td>
<td>950</td>
<td>3.51</td>
</tr>
<tr>
<td>D</td>
<td>10,2.5,5</td>
<td>210,52,105</td>
<td>248</td>
<td>828</td>
<td>3.14</td>
</tr>
<tr>
<td>E</td>
<td>8,3,9</td>
<td>168,63,189</td>
<td>247.5</td>
<td>1060</td>
<td>3.92</td>
</tr>
<tr>
<td>F</td>
<td>6,6,3.5</td>
<td>126,126,73</td>
<td>249.5</td>
<td>212</td>
<td>0.78</td>
</tr>
<tr>
<td>G</td>
<td>12,4.5,2.5</td>
<td>252,94,52</td>
<td>248.5</td>
<td>636</td>
<td>2.35</td>
</tr>
<tr>
<td>H</td>
<td>17,9,4.5</td>
<td>357,189,94</td>
<td>249</td>
<td>424</td>
<td>1.57</td>
</tr>
<tr>
<td>I</td>
<td>2.5,2,2</td>
<td>52,42,42</td>
<td>248.5</td>
<td>636</td>
<td>2.35</td>
</tr>
<tr>
<td>J</td>
<td>1,2,1.5</td>
<td>21,42,32</td>
<td>246.5</td>
<td>1484</td>
<td>5.49</td>
</tr>
<tr>
<td>K</td>
<td>1.5,2,2.5</td>
<td>32,42,52</td>
<td>246.5</td>
<td>1484</td>
<td>5.49</td>
</tr>
</tbody>
</table>

Extinction distance at $w = 0.0$ is 270A

Foil width = 2120A = $7.85\xi$

Table 6.2 Analysis of bubbles labelled in figure 6.3
Fig. 6.3(1) Stereo plot of data from micrograph including area shown in fig. 6.3
as references for comparison with simulated bubble images. The similarity between the internal contrast lines seen in figures 6.2 and 6.3 is very clear. The origin of these internal contrast lines has been explained by Goodman and McLean (110) who investigated this type of contrast from MgO cubes near a \(\langle111\rangle\) orientation. A similar set of images was obtained using a \(\langle100\rangle\) orientation in order to facilitate measurement of the error involved in pore size estimation.

Table 6.3 shows the sets of measurements taken from various fringes using a simulated image of a cubic pore of side 250A. In the table the measurements are given in cm's as this is a convenient unit to use. An example of this series is shown as figure 6.4 where the defocussing distance is 0.6\(\mu\)m. This value of defocus was used to obtain all other defocussed images in this report. The differing background contrast in the three images in figure 6.4 is due to the grey level shading routine which was being used at that stage of the work. More sophisticated techniques were used to obtain the other simulated images.

Foreman et al. (107) have recently independently carried out a similar study. They conclude that the most accurate size measurements are obtained using the centre of the first dark ring surrounding the pore with the microscope underfocussed. The pore sizes measured using the centre of the first dark ring and that using the centre of the first light ring surrounding the pore are plotted as figure 6.5. It is clear from the figure that Foreman's result has been confirmed by this work. Computations were also carried out to confirm that the position of the centre of this fringe is insensitive to varying \(w\), defect depth, extinction distance and the other parameters involved in the simulation.
### Table 6.3 Hole size measurement using various fringes.

<table>
<thead>
<tr>
<th>Defocus/μm</th>
<th>First White Fringe</th>
<th>First Dark Fringe</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>inside</td>
<td>outside</td>
</tr>
<tr>
<td>+1.2</td>
<td>5.0</td>
<td>6.1</td>
</tr>
<tr>
<td>+1.0</td>
<td>5.0</td>
<td>6.2</td>
</tr>
<tr>
<td>+0.8</td>
<td>5.3</td>
<td>6.3</td>
</tr>
<tr>
<td>+0.6</td>
<td>5.4</td>
<td>6.5</td>
</tr>
<tr>
<td>+0.4</td>
<td>5.7</td>
<td>6.6</td>
</tr>
<tr>
<td>+0.2</td>
<td>6.0</td>
<td>6.8</td>
</tr>
<tr>
<td>-0.2</td>
<td>7.1</td>
<td>8.4</td>
</tr>
<tr>
<td>-0.4</td>
<td>7.4</td>
<td>8.5</td>
</tr>
<tr>
<td>-0.6</td>
<td>7.3</td>
<td>8.9</td>
</tr>
<tr>
<td>-0.8</td>
<td>7.4</td>
<td>9.1</td>
</tr>
<tr>
<td>-1.0</td>
<td>7.6</td>
<td>9.3</td>
</tr>
<tr>
<td>-1.2</td>
<td>7.8</td>
<td>9.8</td>
</tr>
</tbody>
</table>

At the in focus plane the measurement is 7.3cms.
Fig. 6.4 Through focal series, <100> orientation.

true pore size is 7.3cm.

Fig. 6.5 Hole size measurement
Figure 6.6 shows how the image changes with foil orientation for a cubic bubble of side 250Å which is situated in the centre of the foil. Comparison with experimental images in similar orientations can be made using figure 6.3, in which a <111> foil orientation is evident and figure 6.18 in which a <100> is displayed. The same model was used to compute figure 6.7 in which the effect of varying w is displayed. As expected the image becomes less crisp when w is increased. This corresponds to moving away from strong diffraction conditions to weak, many-beam type conditions. The image was found to be insensitive to changes in the w value of ±0.1. This is about the accuracy to which w can be measured. Figure 6.8 illustrates the image variation when the defect depth is varied. In the figure the defect depths are in extinction distance units. The overall characteristics of the image remain stable despite the depth changes but the contrast of the central 'dot' in the defocused images does change. Figure 6.3 shows bubbles at a wide range of depths, see table 6.2, all of which show similar contrast features. The bubble marked 'b' in the figure may show dark central dot contrast but as this is the only one to exhibit this contrast feature no conclusions can be drawn. Cube size is varied in figure 6.9 from which it can be seen that the image is insensitive to changes in this parameter in the range displayed. For hole sizes of less than one extinction distance the image does change with the contrast of the central 'dot' being white at one extinction distance and black at other sizes. For hole sizes greater than 0.75 extinction distances the internal line contrast becomes clear. The bubble marked 'f' in figure 6.3 is one of the smallest with clear internal line contrast shown in the micrograph. From table 6.2 the size of this bubble is about 125Å*125Å*70Å. Using the extinction distance for the appropriate reflection, i.e. 270Å implies that the
Foil orientation variation, cubic hole of side 250A in the centre of the foil.
Fig. 6.7 Deviation from the Bragg position varied, cubic pore in the foil centre.
Fig. 6.8 Pore depth variation using a cubic pore and deviation parameter = 1.0.
a. cube side = 1.25
b. cube side = 1.5
c. cube side = 2.0

Pore size variation

Fig. 6.9 Pore size variation, cubic pore at the foil centre and deviation parameter = 1.0.
bubble is about $0.5\cdot 0.5\cdot 0.25$ in size. However it must be remembered that although the precise value of $w$ used to obtain the micrograph is not known, it is certainly quite large. If $w=1$ the effective extinction distance drops from 270A to 190A, using equation 2.16, such that the bubble dimensions are now $0.65\cdot 0.65\cdot 0.36$ expressed in extinction distance units. Thus it may only be deduced from the experimental images that bubbles must be a certain size in order to exhibit internal line contrast and that a size limitation of $0.75\cdot 1$ may be of the correct order. All the previous simulated images have been of cubic pores. Figure 6.10 is of brick shaped pores viewed in different orientations. Strong similarities between the images in figure 6.10(a) and some of those in figure 6.3 are very clear.

Figures 6.2 to 6.10 show that all the parameters discussed in chapter 5 can be altered in the simulation program. In the remainder of this chapter the strain field surrounding the defect will be included.

6.2.2. Strain contrast images

The simplest bubble shape and strain field combination is to consider both as being spherical. Before moving to more realistic models an example will be given using these approximations. It was suggested in section 2.2 that the direction of the black-white lobe contrast should be opposite for voids and overpressurised bubbles. Figure 6.11 shows this effect for a spherical pore of 50A diameter. Using this observation in conjunction with known reference figures, such as figure 2.5, may allow differentiation between voids and overpressurised bubbles when the defect depth can be measured.
Fig. 6.10 Brick shaped pores at foil centre with deviation parameter = 1.0.
a. gas pressure $= 1.1 \times 10^7$ Nm$^{-2}$

b. gas pressure $= 1.1 \times 10^9$ Nm$^{-2}$

equilibrium pressure $= 1.1 \times 10^9$ Nm$^{-2}$

Fig. 6.11 Spherical strain field surrounding spherical pores.
BUB. SIZE = 50.00
BEAM ALONG (111)
DEPTH = 125.00 ANGSTROM
THICKNESS = 1044.00 ANGSTROM
\( \omega \) (DEVM) = -.10
DIFF. VEC = (11)
NPTS = 127
INTERP = 2.

DEFOCUS = 0 A
CS = 1.8E-01 A
WAVELENGTH = 3.7E-02 A
Figure 6.12 is a series of images of a large cubic pore containing a range of pressures using a spherical approximation for the strain field. The pore sides are 200A with \( w = 0.0 \) and all images are in focus. The pressure values used are given in the figure. In figure 6.12(1) the same series is shown with the diffraction vector changed from \(<220>\) in 6.12 to \(<200>\) in 6.12(1). There is a limit to the gas pressure that can be contained in a bubble before interstitial dislocation loops are punched from the bubble. For spherical bubbles this limit is of the order of \( Gb/r \) where \( G \) is the shear modulus\(^{114}\). When considering large faceted pores this may be a rather naive assumption as the energy required to nucleate the loop may be different at the edges and corners of the faceted pore. Unfortunately in the absence of an alternative in the literature this assumption has been used. It must be noted that bubbles may be able to contain a higher maximum pressure than suggested by this formula. A cubic bubble of side 200A is approximately equivalent, using a volume criterion, to a spherical bubble of radius 120A. Thus the maximum pressure that such a bubble can contain is of the order of \( 0.8GNm^{-2} \). In figure 6.12 strain contrast is just visible when the pressure is equal to \( 0.5GNm^{-2} \). As this contrast is is probably too weak to appear in a real micrograph these figures suggest that the strain field becomes visible when the gas pressure contained is between \( 0.5GNm^{-2} \) and \( 0.75GNm^{-2} \). A white fringe can be seen inside the black lobe in figure 6.12(a). This effect will be discussed with reference to the images obtained using the Green's function formulation. The same bubble shape and strain field configuration is shown in figure 6.13. This figure shows the bubble containing a very high gas pressure in different orientations using different \( g \) vectors.
Pressure units are 10 Nm

Spherical Strain Field

Fig.6.12 Cubic pore modelled with a spherical surrounding strain field.
Fig. 6.12(1) Pressure variation using the spherical approximation to calculate the strain field with \( g = <200> \).
Spherical calc.

pm29          pm30

g=220          g=-220
hk.l=001       hk.l=111
pres=.10E11Nm⁻²
w=0.30
xₜₘₚₙ, yₜₘₚₙ=630A

Fig.6.13 Cubic pore modelled with a spherical surrounding strain field.
Further investigation of the pressure necessary to allow the strain field to be seen was carried out using the Green's function technique discussed in section 5.4.1. The results for a range of pressures at $\omega = 0.0$ are presented as figure 6.14. Using this approximation the strain field becomes visible with gas pressure between $0.5 \text{GNm}^{-2}$ and $0.75 \text{GNm}^{-2}$. Comparing figures 6.12 and 6.14 it is clear that the strain field contrast is larger using the Green's function approximation. This is consistent with Meissner's comment mentioned previously. Profiles of the line of maximum extent of the strain contrast were plotted from which table 6.3 was compiled. The range was measured assuming that a 10% change from the background contrast level is visible. Figure 6.15 is an example of the profiles and figure 6.16 are plots of the data in table 6.3. The graphs suggest that the rate of increase in the range of the strain field reduces at high pressures. This means that we can not use the extent of the strain contrast to relate the gas pressures contained in different pores. In figures 6.14(a) to (f) the white fringe also seen in figure 6.12 is clear. In 6.14(f) the pressure is such that the lattice planes at the edge of the pore are tilted to the Bragg position. A possible explanation is that the pressure is increased in (e) to a point where the planes are now tilted beyond the Bragg position so that a light fringe occurs. Further from the edge of the pore the planes are less deformed such that they are again at the Bragg position and a black fringe is again visible. Figures 6.14(a) to (d) show a black fringe inside this white fringe. This implies that some of the lattice atoms have been displaced by more than one lattice parameter and that another plane at the Bragg position has been formed. Fringe structures such as these are not seen experimentally as the pressures necessary for them to occur are much
pressure in Nm * 10
a. 10   f. 0.75
b. 7.5  g. 0.5
in all images
w = 0.0
c. 5.0  h. 0.25
d. 2.5  i. 0.1
e. 1.0

Fig. 6.14 Varying pressure in bubble

Fig. 6.14 Cubic pore, strain field calculated using the finite element Green's function technique.
Table 6.4 Range of strain contrast at various pressures.

<table>
<thead>
<tr>
<th>Pressure values  (/GNm^{-2})</th>
<th>range of strain field/(\text{cms.})</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.10</td>
<td>3.4</td>
</tr>
<tr>
<td>0.25</td>
<td>3.4</td>
</tr>
<tr>
<td>0.50</td>
<td>3.4</td>
</tr>
<tr>
<td>0.75</td>
<td>3.4</td>
</tr>
<tr>
<td>1.00</td>
<td>3.7</td>
</tr>
<tr>
<td>2.50</td>
<td>4.4</td>
</tr>
<tr>
<td>5.00</td>
<td>7.4</td>
</tr>
<tr>
<td>7.50</td>
<td>8.5</td>
</tr>
<tr>
<td>10.0</td>
<td>9.4</td>
</tr>
</tbody>
</table>
Pressure values are $10^9$ Nm$^{-2}$.

Fig 6:15 Strain contrast profiles.
Fig 6.16 Maximum range of strain contrast images.
greater than that required for dislocation loops to be punched from the bubbles.

The bubble illustrated in fig. 6.14(f) was chosen to investigate the effect of varying \( w \) on the strain field contrast with the results shown as figure 6.17. It is interesting to note that a large contrast change takes place between \( w = 1.0 \) and \( w = 2.0 \). The same sequence of bubbles are shown as figure 6.17(1) with the images underfocussed by 0.6/fm. Defocusing the microscope makes the strain contrast invisible in all but the lowest \( w \) image displayed at this value of pressure. A similar procedure was applied to the bubbles shown in figure 6.12(1) from which it was clear that a similar effect occurs. The strain field only became visible at a pressure of 56Nm\(^{-2}\) when the image was defocussed at this value of \( w \). The reason for this effect lies in Fourier transform theory. The in focus image, when the hole is viewed in a \(<100>\), is effectively a rect function which becomes a sinc function after the two dimensional Fourier transform has been taken. As the edges of the rect function get rounded off by the strain field of the defect the amplitude of the ringing in the sinc is reduced. The extreme case of this is the Gaussian function whose transform is another Gaussian. Thus small changes in the contrast due to the strain field get swamped by the transfer function contrast changes.

A series of experimental images at varying \( w \) and \( \theta \) values are presented as figure 6.18. The material being imaged is niobium/1% zirconium containing helium bubbles, the magnification is 600K, and the microscope is defocussed slightly. The micrographs shown as (a) and (b) have the same \( \theta \) vector but are at different values of \( w \). Unfortunately the precise \( w \) values are not known as Kikuchi lines
Fig. 6.17  Cubic hole, strain field calculated using the Green's function method, varying the deviation parameter.

Fig 6.17  Varying w value

\[ U_J = 0.5 \]
\[ b - u_j = 1.0 \]
\[ c - U_J = 2.0 \]
\[ d - U_J = 3.0 \]
Fig. 6.17(1) The same series as shown in Fig. 6.17 but with the images defocussed.
were not present in the diffraction patterns. A rough idea of the $w$ value can be gained from the position of the weak extinction contour seen in the images. The bubble labelled 'A' exhibits very small strain contrast in (a) when $w$ is quite large but distinct contrast when the contour moves across it in (b). This corresponds to a very small local value of $w$. In figures (c) and (d) the contours are in similar positions but the $g$ vectors are different. It is likely that the indexed value of $g$ in (d) is incorrect as the contrast exhibited is typical of a small $g$. It is possible that the diffraction pattern corresponding to this micrograph was not taken of the specific region shown in the figure. Local bending of the foil would result in variation of the operating reflection. Figure (c) is typical of a high $g$, low $w$ reflection while (d) suggests low $g$, high $w$ diffraction conditions. All these experimental images are obtained with the foil in a $<100>$ orientation which may be compared with figure 6.4.

The effect of increasing the $g$ vector is illustrated by figure 6.19 in which 'in focus' images are displayed of a cubic pore of side 200Å containing gas at a pressure of 5GNm$^{-2}$. It is clear from this series that although the extent of the strain field is increased with increasing $g$ it still remains limited. This agrees with the accepted wisdom of using as large a $g$ as possible when imaging strain fields. Unfortunately this ideal can not be realised in the microscope as it is almost impossible to get good, two-beam conditions when using a very large $g$. Typically a $g$ of about $<310>$ is used as a compromise between these two conditions.
Fig. 6.18  Helium bubbles in niobium/1% zirconium which had been annealed for 8 hours at 1250°C. The images are at various υ values.
It is interesting to note that the strain fields exhibited in the experimental images are not symmetrical. The bubble labelled 'A' in figure 6.18 has a small lobe apparently coming from one of the corners. The calculated strain fields shown in figure 6.12 to 6.17 also display asymmetry. A possible reason for the experimental asymmetry is that the foil being imaged may have been deformed such that the local value of \( w \) was different on either side of the pore. Anisotropy can not be used to explain the asymmetry in the computed images as it is implicit in the model that the matrix is isotropic. The bubble labelled 'B' in figure 6.18 shows interesting contrast in this series of micrographs. In (a) the weak contour is to one side of the bubble and a strain lobe is visible on this side if the bubble. In (b) the contour has moved across the bubble which corresponds to changing the sign of \( w \) and the strain lobe has now also swapped position. Figure (c) shows the bubble with strain contrast appearing on both side of the bubble and also inside the projection of the bubble while in (d) no strain contrast is exhibited. Although most of the images in figure 6.18 do not show strain contrast within their projections this type of image is often seen in the microscope(115).

The analytic Green's function method has not yet yielded presentable results as images obtained by this method have always included a very large contrast change along the projected edges of the pore. The magnitude of this contrast change may have swamped the derived strain contrast although this is likely to be incorrect as the lattice displacement does not fall off with distance from the pore as it should. This point will be discussed in the appendix.
Hole is at the foil centre
w = 0.0
Pressure = 5. GPa
Diffraction vectors are:
110  200  220
310  400  420
510

Fig. 6.19 Varying the operating reflection
CHAPTER SEVEN

CONCLUSIONS
The aim of this project was twofold.

(1) To produce and image voids in a high voltage electron microscope in order to validate Fisher's technique by calculating the vacancy migration energy and point defect bias in nickel.

(2) To write an image simulation program which would allow various morphologies for cavities and their strain fields to be included.

The first aim has been achieved with the vacancy migration energy being calculated to be $1.15(\pm0.05)eV$ and the interstitial bias to be 6.5%. The possibility that voids may also have a stronger attraction for interstitials than vacancies has also been investigated with the result that the void bias has been calculated to be 0.5% in nickel. Scope for future work in this field is very great as the method can be applied to many other materials.

The second aim has also been achieved in that a portable computer program has been written which can incorporate realistic pore shapes and strain fields. This has been used to deduce that the most accurate pore size measurements are made with the microscope underfocussed and using the first dark ring surrounding the pore. Image reversal between overpressurised bubbles and voids has been displayed along with the effects of varying the parameters involved in image simulation. Computations have been performed on large faceted pores with their strain fields being modelled as both spherically symmetric and non-spherically symmetric. In the latter case results have been presented using a finite element method and an analytical approach has been attempted.
Results have been yielded by the strain field simulations which correlate well with those observed experimentally. The extent of strain field contrast, which is always fairly small in the microscope, has been shown to be very limited in the computed images. It has been demonstrated that increasing the magnitude of the diffraction vector does not produce a large increase in the extent of the strain contrast. Experimental strain field images are often taken with the microscope defocussed to clarify the defect shape. It is clear from this work that defocussing the microscope results in a reduction in the size, or even the loss, of the strain contrast. The results also suggest that the pressure range accessible by interpretation of strain images is only a small part of the range of interest.

The primary aim of future work on this topic would be to remove the bugs in the analytic approach routines as this method is nearly ten times faster than the finite element method. The similarity of the images computed with spherical strain fields to those obtained experimentally suggests that this may be a reasonable method. Another advantage of this technique is speed as it is about sixty times faster than even the analytical Green's function method. However there are disadvantages with this approximation for cubic or brick shaped pores as assigning a radius equivalent to the pore dimensions is clearly questionable. Another problem with this method is that no allowance could be made for anisotropy in the matrix material. The finite element method does offer scope for future work as it could be modified to incorporate both anisotropy in the matrix material and the defect Green's function. Although this technique is rather cumbersome the programs are presently being transferred, by Dr.
K. Ahmad to the Cray computer at the Rutherford laboratory which should result in a large reduction in the computing time. Modelling the pore as six sheets of force acting normal to the faces has its limitations. A better model might be to include an array of forces acting along the $<111>$ as this would lead to increased displacement at the corners to correspond to the apparent strain contrast often seen in the microscope. No allowance is made for any deformation that may occur to the pore faces in this model. It may be possible to design a recursive technique whereby the displacement calculated at one point on the hole face is used to modify the origin of the force in the next element. These modifications have not been made in view of the restrictions on computation time.

Applications of a system which could allow estimation of the pressure within pores occur in many fields with the nuclear power industry being the main one. Malen and Bullough in 1971 stated that precise representation of a faceted pore would probably be impossibly complicated. The truth of their statement has been modified by this work but despite this it still remains a fair comment.
It was stated in chapter 5 that in order to arrive at an analytical solution for the strain fields using the Green's function method the only integral required is of the form:

\[ J'(x_1, x_2, x_3, a, b, c) = \int_{-b}^{b} \int_{-c}^{c} dx_2' dx_3' \left[ \frac{1}{(x_1 - a)^2 + (x_2 - x_2')^2 + (x_3 - x_3')^2} \right] \]

This integral can be written as:

\[ J = \int dy \int dz \left[ \frac{1}{(u-a)^2 + (v-y)^2 + (w-z)^2} \right] \]

where \( u, v, w = x_1, x_2, x_3 \)

and \( a, y, z = x_1', x_2', x_3' \).

Let \( w - z = -t \)

and \( v - y = -s \)

then \( dz = dt \)

and \( J = \int ds \int dt \left[ \frac{1}{(u-a)^2 + s^2 + t^2} \right] \)

Using \( \int dx / [a^2 + x^2]^{1/2} = \ln[(x + \sqrt{x^2 + a^2})/a] \)

we obtain:

\[ J = \int ds \int dt \ln[t + (u-a)^2 + s^2 + t^2] \]

Thus substituting the integral limits we have:

\[ J = \int ds \int dt \ln[c-w + (u-a)^2 + (c-w)^2 + s^2 + t^2] - \ln[-c-w + (u-a)^2 + (c+w)^2 + s^2 + t^2] \]

Referring to the symbols used in chapter 5 this integral may be interpreted as:

\[ J = J_1(c) - J_1(-c) \]

Let us consider \( J_1(c) \).

\[ J_1(c) = \int ds \ln[c-w + (u-a)^2 + (c-w)^2 + s^2] \]

Say \( x = s \)

\( a = c-w \)

\( b^2 = (u-a)^2 + (c-w)^2 \).

The integral is then of the form:
\[ \int dx \ln[a + (b^2 + x^2)^{1/2}] \]

where the limits of integration from now on are assumed.

This may be integrated by parts to give:

\[ x \cdot \ln[a + (b^2 + x^2)^{1/2}] - \int x^2 dx / [a + (b^2 + x^2)^{1/2}] (b + x^{1/2}) \]

Multiplying the denominator and numerator by \(a - (b^2 - x^2)^{1/2}\) the second term in this expression becomes:

\[ - \int x^2 [a - (b^2 + x^2)^{1/2}] dx / [a^2 - b^2 - x^2] (b + x^{1/2}) \]

Now:

\[
\int x^2 [a - (b^2 + x^2)^{1/2}] dx = - \int \frac{a - (b^2 + x^2)^{1/2}}{(b^2 + x^2)^{1/2}} \int \frac{a - (b^2 + x^2)^{1/2}}{(a^2 - b^2 - x^2) (b^2 + x^2)^{1/2}} dx
\]

Thus the original integral has now reduced to:

\[ x \cdot \ln[a + (b^2 + x^2)^{1/2}] + \int \frac{a - (b^2 + x^2)^{1/2}}{(b^2 + x^2)^{1/2}} dx - \int \frac{a - (b^2 + x^2)^{1/2}}{(a^2 - b^2 - x^2) (b^2 + x^2)^{1/2}} dx \]

This may be written:

\[ x \cdot \ln[a + (b^2 + x^2)^{1/2}] + a \frac{dx}{[b^2 + x^2]^{1/2}} - x - (a - b) \int \frac{a - (b^2 + x^2)^{1/2}}{(a^2 - b^2 - x^2) (b^2 + x^2)^{1/2}} dx \]

which then becomes:

\[ x \cdot \ln[a + (b^2 + x^2)^{1/2}] + a \frac{dx}{[b^2 + x^2]^{1/2}} - x - (a - b) \int \frac{a - (b^2 + x^2)^{1/2}}{(a^2 - b^2 - x^2) (b^2 + x^2)^{1/2}} dx \]

We shall now consider the last term, which can be written as:

\[ (a - b) \int \frac{dx}{[a - b - x^2] (b^2 + x^2)^{1/2}} - \int \frac{dx}{[a^2 - b^2 - x^2]} \]

Using \((a - b - x^2)^{-1/2} = \left[ (a^2 - b^2) + x \right]^{-1} + \left[ (a^2 - b^2) + x \right]^{-1} \right] \cdot 1/2 \cdot \frac{1}{(a - b)}^{1/2} \]

the last term becomes:

\[
\frac{1}{2(a^2 - b^2)^{1/2}} \int \frac{dx}{[a^2 - b^2 - x]^{1/2} + x} + \frac{1}{[a^2 - b^2 + x]^{1/2}} \int dx.
\]

This then becomes \(0.5 \cdot \frac{1}{(a^2 - b^2)^{1/2}} \ln \left[ (a^2 - b^2)^{1/2} + x \right] / (a^2 - b^2)^{1/2} - x \].

The only remaining term is of the form:

\[ a(a - b) \int \frac{dx}{[a - b - x] (b^2 + x^2)^{1/2}} \]

Using the relation given above for \(1 / (a - b - x^2)\) we have:

\[
\int \frac{dx}{[a - b - x] (b^2 + x^2)^{1/2}} = \frac{1}{2(a^2 - b^2)^{1/2}} \int \frac{dx}{[a^2 - b^2 - x] (b^2 + x^2)^{1/2}} + \int \frac{dx}{[a^2 - b^2 + x] (b^2 + x^2)^{1/2}}
\]

Consider the term \(\int dx / (a^2 - b^2 + x) (b^2 + x^2)^{1/2}\).
Letting \( p = (a^2 - b^2)^{\frac{1}{2}} \) this becomes \( \int dx/(p+x)(b+x)^{\frac{1}{2}} \).

Now let \( t = p + x \)

\[
dt = dx
\]

then the integral can be expressed as:

\[
\int dt/t(b^2 + (t-p)^{\frac{1}{2}})
\]

Now using the substitution \( t = 1/y \) and thus \( dt = -dy/y^2 \)

we have the integral in the form:

\[
- \int y dy/ y^2[b^2+(1/y - p)^{\frac{1}{2}}]
\]

This can be rewritten as:

\[
- \int dy/ [b^2y^2+(1-py)^{\frac{1}{2}}]
\]

which in turn becomes:

\[
- \int dy/ [ y^2[b^2+p^2] -2yp +1]^{\frac{1}{2}}
\]

This integral is of the form:

\[
\int dZ/ [AZ^2 + BZ + C]^{\frac{1}{2}}
\]

Now

\[
\int dZ/ [AZ^2 + BZ + C]^{\frac{1}{2}} = \int dZ/[[AZ^2 +B/2A] + C - B^2/4A]^{\frac{1}{2}}
\]

Let \( U = A^{\frac{1}{2}}Z + B/2A^{\frac{1}{2}} \) and thus \( dU = A^{\frac{1}{2}}dZ \).

The integral is now:

\[
A^{-\frac{1}{2}} \int dU/ (U^2 + C - B^2/4A)^{\frac{1}{2}}
\]

which is:

\[
A^{\frac{1}{2}} \ln[U + (U^2 + C - B^2/4A)]^{\frac{1}{2}}
\]

Thus all the terms in the original integral have been integrated and it only remains to substitute the variables and limits of integration to solve the problem. As the complete result is very lengthy I shall only give as an example the substitution into the last term from which the complex nature of the solution can be seen. This term becomes:

\[
-\frac{1}{2} \ln[(c-w)x' - [-(c-w)]^{\frac{1}{2}} + \left[ (c-w)x' - (c-w) \right]^{\frac{1}{2}} + (c-w)]^{\frac{1}{2}}
\]

where \( x' \) represents the integration limits, in this case \(-b-v\) and \( b-v\).
Differences between the form of this solution and the finite element method (FEM) are immediately evident. In the FEM we are always dividing by \( R \), where \( R = \left[ (x_i - x'_i)^2 + (x_j - x'_j)^2 + (x_k - x'_k)^2 \right]^{\frac{1}{2}} \), such that problems with dividing by very small numbers only occur when all of \( (x - x') \), \( (x - x') \), \( (x - x') \) are small. This condition can only hold at the eight corners of a brick-shaped pore and so can be easily incorporated. Using the analytical technique we are dividing by individual terms such as \( (c-w) \) in the above equation. Small values for these terms occur along the projected edges of the pore and extend out to the complete range of the picture which makes this effect rather harder to deal with. The equations in the FEM have a definite \( 1/R \) fall off for the displacement whereas those arrived at by the analytical technique have \( \ln(R) \) appearing in them, which is a similar form to that obtained by Li(116). It must be remembered that although \( \ln(R) \) increases with \( R \) we are taking \( \ln(R-x) - \ln(R'-x) \) when considering the displacements due to opposite faces. As \( R \) and \( R' \) increase relative to the pore size, \( x \), the \( R \) terms dominate and the factor \( R/R' \) tends to unity such that log of this term reduces. Thus the lattice displacements do reduce as the distance from the hole faces increases.
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