Determination of the Number of Unique Configurations of Unrelaxed Clusters in Crystal Lattices*

A. P. Brody and M. Meshii
Department of Materials Science and Materials Research Center, Northwestern University, Evanston, Illinois

(Received 1 July 1968)

A procedure, applicable for use on a computer, is presented which permits the determination of the number and arrangement of unique n-component unrelaxed clusters for cubic lattices in x-y-z space. The configurations thus obtained are listed for the fcc, bcc, and simple-cubic lattices. The present results apply to clusters which are composed of either vacancies and/or substitutional impurities.

THE crystallography of elementary point defects is simple and can be determined by visual inspection without any difficulty. In considering multiple defects, however, the complexity involved in the determination of the unique configurations rapidly increases with an increase in the number of components of which the cluster is composed, thus vitiating the usefulness of the visual inspection procedure. The present paper describes a procedure, amenable for use on a computer, which overcomes this difficulty associated with the large component clusters. The method is designed for use with cubic lattices because this permits a systematic investigation of all the possible interlattice relationships to be easily investigated and compared.

An example in which such information is important occurs during the course of point-defect studies. In that case, the necessity of determining every unique configuration for an n-component cluster (a cluster consisting of n elementary defects) without duplication or omission is a requirement if energy and entropy calculations of multiple defects as well as an analysis of the interactions between defects is to be carried out.

Although the clusters are assumed to be vacancy clusters in this presentation, the calculations are equally valid for clusters consisting of only substitutional solute clusters, or those containing both such species. That this is so follows from the fact that only lattice sites are considered to be potential resident positions. Thus for unrelaxed clusters the nature of the defect occupying a site is not significant to the calculation.

A cluster will be defined as consisting of n elementary defects, each one of which has at a minimum at least one first nearest neighbor (f.n.n.) bond with any other component of the cluster. By a unique configuration, we shall mean the ensemble of all configurations equivalent under the symmetry operations of the lattice translations and rotations.

The procedure used will first be described and then an example as to how it is applied to a particular case will be presented.

The program is cyclic in nature and can be basically regarded as being analogous to the development of a family tree of relationships among the configurations of n-component clusters in a given lattice. The fcc lattice will be considered below. There is only one unique configuration of a two-component cluster (divacancy) in a fcc [this is also true for the bcc and simple cubic (sc), respectively] and this species was taken as the starting point of the calculations. The two-component cluster was positioned in x-y-z space, and each of the 12 vectors representing the 12 nearest-neighbor translations [e.g., $(\frac{1}{2},\frac{1}{2},0)$, $(-\frac{1}{2},0,\frac{1}{2})$, etc.] was in turn added to each of the two original sites of the divacancy. Any resultant new sites, so determined, which were found to represent any of the two original positions were not considered to be potentially new positions from which possible new configurations might be generated. In addition, only unique f.n.n. sites, with respect to the original cluster, were considered.

The process of the characterization of the clusters was then pursued in three steps. In the first step, the absolute values of the distances between both the vectorally produced new sites and the original sites and between the original sites themselves were determined. Secondly, these distances were then ordered in a descending manner with respect to their absolute values for a given potential $(n+1)$-component cluster. The third step consisted of determining whether the sum of the differences (of a one-one comparison, in an ordered manner) between the absolute values of the distances within each potential new cluster was different from zero when compared to the ordered distances within the other potential new clusters. If this indeed was the

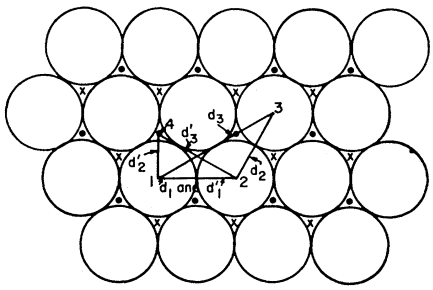

FIG. 1. The (111) plane of the fcc lattice and the original position of the divacancy at sites 1 and 2, with two of their possible 18 unique f.n.n. at sites 3 and 4.

* Research supported by the Advanced Research Projects Agency of the Department of Defense through the Northwestern University Materials Research Center.

TABLE I. CDC 6400 computer execution times necessary to obtain the unique clusters in the three lattices. Times are in seconds.

Lattices	n	CP	PP
sc	6	502	200
bcc	5	8	85
fcc	5	25	195

TABLE II. Number of unique f.n.n. clusters.

$n=$	2	3	4	5	6
fcc	1	4	19	109	...
bcc	1	3	12	51	...
sc	1	2	7	21	99

situation, then the $(n+1)$-component cluster so compared to all the other $(n+1)$-component clusters was unique with respect to the configurational relationships among its components. The unique $(n+1)$-component clusters so obtained were then stored and each of them acted as a starting point for the determination of unique $(n+2)$-component configurations. The three-step comparison test was then repeated, in a manner similar to that described above, for all of the $(n+2)$-component clusters after their f.n.n. positions had been defined through the use of the vectoral addition procedure. With such a technique, the unique $(n+3)$-component clusters were thus determined. The cycle was then repeated as many times as necessary until all the required unique $(n+k)$-component clusters were obtained.

As an example, consider the following: Figure 1 represents the (111) plane of an fcc lattice in which the

TABLE III. Unique unrelaxed n-component configurations.[a,b]

fcc		fcc	
	$n=2$		010, $(-\frac{1}{2})$ $(-\frac{1}{2})$ $\bar{1}$
	000, $(-\frac{1}{2})$ $\frac{1}{2}$ 0	8.	000, $(-\frac{1}{2})$ $\frac{1}{2}$ 0, $\bar{1}$ $\frac{1}{2}$ $(-\frac{1}{2})$, $(-\frac{3}{2})$ 1 $(-\frac{1}{2})$
	$n=3$		$\bar{1}0\bar{1}$, 0 $(-\frac{1}{2})$ $(-\frac{1}{2})$, $\bar{2}$ $\frac{1}{2}$ $(-\frac{1}{2})$, $\bar{2}1\bar{1}$
1.	000, $(-\frac{1}{2})$ $\frac{1}{2}$ 0	9.	000, $(-\frac{1}{2})$ $\frac{1}{2}$ 0, $\bar{1}$ $\frac{1}{2}$ $(-\frac{1}{2})$, $\bar{1}10$
	$\bar{1}00$, $\frac{1}{2}$ $(-\frac{1}{2})$ 0, $(-\frac{1}{2})$ 0 $(-\frac{1}{2})$,		$\frac{1}{2}$ $\frac{1}{2}$ 0, 0 $\frac{1}{2}$ $\frac{1}{2}$, $\bar{1}0\bar{1}$, $(-\frac{3}{2})$ $\frac{3}{2}$ 0, $(-\frac{3}{2})$ $\frac{1}{2}$ 0
	$\bar{1}$ $\frac{1}{2}$ $(-\frac{1}{2})$	10.	000, $(-\frac{1}{2})$ $\frac{1}{2}$ 0, $\bar{1}$ $\frac{1}{2}$ $(-\frac{1}{2})$, $(-\frac{3}{2})$ 0 $(-\frac{1}{2})$
	$n=4$		$\frac{1}{2}$ 0 $(-\frac{1}{2})$, 0 $(-\frac{1}{2})$ $(-\frac{1}{2})$, $\bar{2}$ $\frac{1}{2}$ $(-\frac{1}{2})$, $\bar{2}0\bar{1}$
1.	000, $(-\frac{1}{2})$ $\frac{1}{2}$ 0, $\bar{1}00$	11.	000, $(-\frac{1}{2})$ $\frac{1}{2}$ 0, $\bar{1}$ $\frac{1}{2}$ $(-\frac{1}{2})$, $(-\frac{1}{2})$ $(-\frac{1}{2})$ 0
	$(-\frac{3}{2})$ $\frac{1}{2}$ 0, $(-\frac{1}{2})$ $(-\frac{1}{2})$ 0		010, $(-\frac{1}{2})$ $\bar{1}$ $\frac{1}{2}$, $\bar{1}0\bar{1}$, $(-\frac{3}{2})$ 0 $(-\frac{1}{2})$
2.	000, $(-\frac{1}{2})$ $\frac{1}{2}$ 0, $\frac{1}{2}$ $(-\frac{1}{2})$ 0		$\frac{1}{2}$ 0 $\frac{1}{2}$, $(-\frac{1}{2})$ $\frac{1}{2}$ $\bar{1}$, $(-\frac{3}{2})$ 1 $(-\frac{1}{2})$, 0$\bar{1}$0
	$(-\frac{1}{2})$ $(-\frac{1}{2})$ 0, $\bar{1}00$, $1\bar{1}0$		$(-\frac{3}{2})$ $\frac{1}{2}$ 0, $(-\frac{1}{2})$ $\bar{1}$ $(-\frac{1}{2})$, $\bar{1}\bar{1}0$, $\bar{1}$ $(-\frac{1}{2})$ $(-\frac{1}{2})$
3.	000, $(-\frac{1}{2})$ $\frac{1}{2}$ 0, $(-\frac{1}{2})$ 0 $(-\frac{1}{2})$	12.	000, $(-\frac{1}{2})$ $\frac{1}{2}$ 0, $\bar{1}$ $\frac{1}{2}$ $(-\frac{1}{2})$, $\bar{1}1\bar{1}$
	$\bar{1}00$, 0 $\frac{1}{2}$ $(-\frac{1}{2})$		$\bar{1}$ $\frac{1}{2}$ $\frac{1}{2}$, $\bar{1}$ $\frac{1}{2}$ $(-\frac{3}{2})$, 0 $(-\frac{1}{2})$ $(-\frac{1}{2})$
4.	000, $(-\frac{1}{2})$ $\frac{1}{2}$ 0, $\bar{1}$ $\frac{1}{2}$ $(-\frac{1}{2})$		$(-\frac{3}{2})$ 0 $(-\frac{1}{2})$, $(-\frac{3}{2})$ 1 $(-\frac{3}{2})$, $(-\frac{1}{2})$ $(-\frac{1}{2})$ 0
	$(-\frac{1}{2})$ 1 $\frac{1}{2}$, $(-\frac{3}{2})$ 1 $(-\frac{1}{2})$, $(-\frac{1}{2})$ $(-\frac{1}{2})$ 0, $(-\frac{1}{2})$ 1 $(-\frac{1}{2})$		$\frac{1}{2}$ 0 $(-\frac{1}{2})$, $\bar{1}0\bar{1}$, $(-\frac{3}{2})$ $\frac{3}{2}$ $\bar{1}$
	$(-\frac{3}{2})$ $\frac{1}{2}$ 0, $\bar{1}10$, $\bar{1}1\bar{1}$, $(-\frac{3}{2})$ $\frac{1}{2}$ $\bar{1}$	13.	000, $(-\frac{1}{2})$ $\frac{1}{2}$ 0, $\bar{1}$ $\frac{1}{2}$ $(-\frac{1}{2})$, 0 $\frac{1}{2}$ $(-\frac{1}{2})$
	0 $(-\frac{1}{2})$ $(-\frac{1}{2})$, $(-\frac{3}{2})$ 0 $(-\frac{1}{2})$, 0 $\frac{1}{2}$ $(-\frac{1}{2})$, $(-\frac{1}{2})$ 0 $(-\frac{1}{2})$		$\frac{1}{2}$ $\frac{1}{2}$ 0, 0 $(-\frac{1}{2})$ $\frac{1}{2}$, $(-\frac{3}{2})$ 1 $(-\frac{1}{2})$, $(-\frac{1}{2})$ $(-\frac{1}{2})$ 0
	$n=5$		0 $\frac{1}{2}$ $\frac{1}{2}$, $\frac{1}{2}$ $\frac{1}{2}$ $\bar{1}$, $\bar{1}10$, $01\bar{1}$
1.	000, $(-\frac{1}{2})$ $\frac{1}{2}$ 0, $\bar{1}00$, $(-\frac{3}{2})$ $\frac{1}{2}$ 0		$(-\frac{3}{2})$ $\frac{1}{2}$ 0, 0 $(-\frac{1}{2})$ $(-\frac{1}{2})$, $(-\frac{3}{2})$ 0 $(-\frac{1}{2})$, $\bar{1}1\bar{1}$
	$\bar{2}00$		$(-\frac{1}{2})$ $\frac{1}{2}$ $\bar{1}$
2.	000, $(-\frac{1}{2})$ $\frac{1}{2}$ 0, $\frac{1}{2}$ $(-\frac{1}{2})$ 0, $(-\frac{1}{2})$ $(-\frac{1}{2})$ 0	14.	000, $(-\frac{1}{2})$ $\frac{1}{2}$ 0, $\bar{1}$ $\frac{1}{2}$ $(-\frac{1}{2})$, $(-\frac{1}{2})$ 1 $(-\frac{1}{2})$
	$\frac{1}{2}$ $\frac{1}{2}$ 0		$\bar{1}$ $\frac{1}{2}$ $\frac{1}{2}$, $\bar{1}0\bar{1}$, $(-\frac{3}{2})$ 0 $(-\frac{1}{2})$
3.	000, $(-\frac{1}{2})$ $\frac{1}{2}$ 0, $\frac{1}{2}$ $(-\frac{1}{2})$ 0, $\bar{1}00$		0 $(-\frac{1}{2})$ $\frac{1}{2}$, $(-\frac{3}{2})$ 1 $(-\frac{1}{2})$, $(-\frac{1}{2})$ $(-\frac{1}{2})$ 0
	$\frac{1}{2}$ $\frac{1}{2}$ 0, $(-\frac{3}{2})$ $(-\frac{1}{2})$ 0, $(-\frac{1}{2})$ $(-\frac{1}{2})$ 0	15.	000, $(-\frac{1}{2})$ $\frac{1}{2}$ 0, $\bar{1}$ $\frac{1}{2}$ $(-\frac{1}{2})$, $(-\frac{3}{2})$ $\frac{1}{2}$ $\bar{1}$
	100, $(-\frac{3}{2})$ $\frac{1}{2}$ 0, 0$\bar{1}$0		$\frac{1}{2}$ $\frac{1}{2}$ 0, $\bar{2}$ $\frac{1}{2}$ $(-\frac{1}{2})$, $(-\frac{1}{2})$ $\frac{1}{2}$ $\bar{1}$, $(-\frac{3}{2})$ 1 $(-\frac{1}{2})$
4.	000, $(-\frac{1}{2})$ $\frac{1}{2}$ 0, $\frac{1}{2}$ $(-\frac{1}{2})$ 0, $1\bar{1}0$		$(-\frac{3}{2})$ 1 $(-\frac{3}{2})$, $\frac{1}{2}$ $(-\frac{1}{2})$ 0, 010, $(-\frac{3}{2})$ $\frac{1}{2}$ 0
	$(-\frac{1}{2})$ $(-\frac{1}{2})$ 0, $\bar{1}00$, $\frac{3}{2}$ $(-\frac{3}{2})$ 0		$\frac{1}{2}$ 0 $(-\frac{1}{2})$, $\bar{2}0\bar{1}$, $\bar{1}1\bar{1}$, $\bar{2}$ $\frac{1}{2}$ $(-\frac{3}{2})$
5.	000, $(-\frac{1}{2})$ $\frac{1}{2}$ 0, $(-\frac{1}{2})$ 0 $(-\frac{1}{2})$, $\bar{1}00$		$\frac{1}{2}$ 0 $\frac{1}{2}$, $\bar{1}$ $\frac{1}{2}$ $\frac{1}{2}$, $(-\frac{3}{2})$ 0 $(-\frac{3}{2})$, $(-\frac{3}{2})$ 0 $(-\frac{1}{2})$
	$(-\frac{1}{2})$ $(-\frac{1}{2})$ 0		0 $\frac{1}{2}$ $(-\frac{1}{2})$, 0 $\frac{1}{2}$ $\frac{1}{2}$, 0 $(-\frac{1}{2})$ $\frac{1}{2}$, 0 $(-\frac{1}{2})$ $(-\frac{1}{2})$
6.	000, $(-\frac{1}{2})$ $\frac{1}{2}$ 0, $\bar{1}$ $\frac{1}{2}$ $(-\frac{1}{2})$, $(-\frac{3}{2})$ $\frac{1}{2}$ 0		$(-\frac{1}{2})$ $(-\frac{1}{2})$ 0, $(-\frac{1}{2})$ 1 $(-\frac{1}{2})$, $(-\frac{1}{2})$ 1 $\frac{1}{2}$, $\bar{1}$ $\frac{1}{2}$ $(-\frac{3}{2})$
	$\frac{1}{2}$ $\frac{1}{2}$ 0, $\bar{2}$ $\frac{1}{2}$ $\frac{1}{2}$, $\bar{2}10$, $\bar{2}$ $\frac{1}{2}$ $(-\frac{1}{2})$		$\bar{2}1\bar{1}$, $\bar{1}00$, $\bar{1}0\bar{1}$
	010, $\bar{1}$ $\frac{1}{2}$ $\frac{1}{2}$, $\bar{2}00$	16.	000, $(-\frac{1}{2})$ $\frac{1}{2}$ 0, $\bar{1}$ $\frac{1}{2}$ $(-\frac{1}{2})$, $(-\frac{1}{2})$ 0 $(-\frac{1}{2})$
7.	000, $(-\frac{1}{2})$ $\frac{1}{2}$ 0, $\bar{1}$ $\frac{1}{2}$ $(-\frac{1}{2})$, 0 $(-\frac{1}{2})$ $(-\frac{1}{2})$		$(-\frac{1}{2})$ $(-\frac{1}{2})$ $\bar{1}$, $(-\frac{3}{2})$ 0 $(-\frac{1}{2})$, 0 $\frac{1}{2}$ $(-\frac{1}{2})$, $\bar{1}0\bar{1}$
			$\bar{1}$ $(-\frac{1}{2})$ $(-\frac{1}{2})$, $\bar{1}1\bar{1}$, $(-\frac{1}{2})$ 1 $(-\frac{1}{2})$, $(-\frac{3}{2})$ $\frac{1}{2}$ $\bar{1}$

[a] The numbered x-y-z coordinates are the starting positions from which the unnumbered sites, given immediately below, were generated. Thus, for example, there are four unique three-component configurations, each one of which contains the sites 000 and $(-\frac{1}{2})$ $\frac{1}{2}$ 0.

[b] A copy of the program written in FORTRAN IV for use on a CDC 6400 series computer is available on request, as are the results obtained for the bcc and sc lattices.

immediate upper and lower planes are given by dots and crosses, respectively. Positions marked 1 and 2 represent an original divacancy, while those given by 3 and 4 are two of the divacancy's unique f.n.n. Let d_1 represent the absolute value of the distance between sites 1 and 2, d_2 that between 2 and 3, and finally d_3 that between positions 3 and 1. (The absolute value of the distances between all original positions and between all new and original positions are thus given.) Since $d_3>d_2=d_1$, these values may be ordered as d_3, d_2, d_1. Now, consider point 4 and its relationship to positions 1 and 2. Again, the values of the absolute distances between the sites involved may be arranged in a descending manner to obtain d_3', d_2', d_1'. After subtracting as follows: $(d_3-d_3')=a$, $(d_2-d_2')=b$, and $(d_1-d_1')=c$, the sum of $a+b+c$ is obtained. If the sum is not equal to zero, then the position represented by the ordered vectors $d_3d_2d_1$ is unique with respect to those positions represented by $d_3'd_2'd_1'$.

The fcc, bcc, and sc lattices were investigated. It was found that the number of unique configurations of a four-component cluster in an fcc is 19 and not 20 as previously reported.[1] An examination of diagrams 7 and 8 of Fig. 9 of Ref. 1 indicates that these two configurations are actually identical in shape though of different orientations with respect to a fixed origin.

The unique configurations listed below are inclusive of a six-component cluster in the sc and a five-component cluster in both the fcc and the bcc. There is no definite limitation as to how far the determination of unique clusters may be carried out using the procedure described above, because the program is cyclic and can, in effect, feed upon itself; however, both the execution time as well as available computer memory space can be expected to place an upper limit on the extent of calculations beyond what is absolutely required on an informational basis.

It can be seen that by using the general outline of the above scheme it is possible to develop a program to determine the geometric constants for a given defect reaction associated with the various unique cluster configurations. The program, however, would be much more complicated than the one described here because it would not be possible in this instance to utilize a general systematic procedure.

The program was executed on a CDC 6400 series computer. The compilation time of the program was approximately 3 sec of central processor (CP) time and 24 sec of peripheral processor (PP) time. The execution times necessary to obtain the results shown below are given in Table I along with the maximum sized clusters found in each instance. The CP and PP times are in seconds. A summary of the results are presented in Table II. In Table III the unique unrelaxed n-component configurations are given for the fcc lattice.

ACKNOWLEDGMENT

The authors would like to thank Professor T. Mori of the Tokyo Institute of Technology for some helpful suggestions.

[1] M. Doyama, in *Lattice Defects in Quenched Metals*, edited by R. M. J. Cotterill *et al.* (Academic Press Inc., New York, 1965).