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A stochastic theory for clustering of quenched-in vacancies-V. Temperature dependence of cluster density

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Abstract. It is known that the density of vacancy loops in quenched aluminium and stacking fault tetrahedra in quenched gold show a saturation for low ageing temperatures. The physical mechanism leading to this effect is not well understood. In this paper we consider a simple model which allows us to obtain the temperaure dependence of total density. The analysis shows that the plateau region arises due to the fact that the number of absorption sites of a cluster is larger than the number of emission sites. The temperature dependence of the average number of vacancies in a cluster and the single vacancy concentration in equilibrium with the clusters are discussed.

Keywords. Clustering; vacancy loops; stacking fault tetrahedra; Master equation; stochastic theory; temperature dependence.

1. Introduction

The present paper is a continuation of our effort to understand the various aspects of clustering. Here, we consider a situation concerning the temperature dependence of cluster density for low ageing temperatures T. It is found that the cluster density $\bar{N}$ saturates for low T. This behaviour is reported for stacking fault tetrahedra in gold (Meshii 1965) and vacancy loops in aluminium (Kiritani 1973 and the references therein). Kiritani (1973) has considered the problem of clustering in detail. His computer results also show this feature. However, he has made no specific attempt to analyse the underlying physical mechanisms leading to this feature. Our own earlier work gave no indication although, we did devote considerable attention to this problem (Ananthakrishna 1979a, b, c, d). We succeeded only in obtaining the temperature dependence of $\bar{N}$ given the dependence of the average size or vice versa.

The experimentally reported results concern the temperature dependence of the asymptotic (large time) cluster density. If one wants to obtain information about this, normally one has to follow the time dependence right from nucleation to the completion of growth. In the first paper of this series (Ananthakrishna 1979a, referred to as I hereafter) we have discussed these two aspects of clustering in a slightly different perspective. In order to emphasize some points, we discuss below nucleation and growth. (Needless to say that there will be some repetition.) Often these two are treated separately, although no such clear distinction exists. The two states of the problem are complementary in the sense that in some respects the problem of nucleation is simpler than the problem of growth but is more difficult in some

other aspects. From the point of view of rate theory, the first stage, namely the nucleation, in principle can be understood by solving the appropriate rate equations on a computer. The simplicity lies in the smaller number of coupled equations one has to solve in comparison with the growth problem, where a very large number of equations have to be solved (see for example Wieberg and Vingsbo 1977). However, if one wants to get an analytical solution of the problem, nucleation may be more difficult since the rate constants are complicated functions of the size of the cluster (through the binding energy). In contrast, during their subsequent growth, the dependence on the size is expected to be simple (due to the fact that the binding energy rapidly saturates). This then in principle allows for coarse-grained rate equations whose number is large but can be handled on a computer. In the case of clustering of quenched-in vacancies, Kiritani (1973) devised a ' coarse ' graining procedure called ' grouping method ' which can be used to follow the problem right from the start to the end of clustering. The essential point is to use different group sizes to take care of rapid changes in binding energies for small sizes. Hayns (1976) has extended it to the irradiated situation and concludes that it is perhaps the only method that affords a way of following the time development continuously. Thus there is a need for any analytical approach to the problem of clustering.

The object of this paper is to present a straightforward calculation which tempera- to recognise the cause for this maximum and derive an expression for the allows us ture dependence of $\bar{N}$. *The method used allows us to calculate a stationary solution for the distribution without a need to follow the time dependence continuously* (as is normally required). We will confine our attention to the formation of vacancy loops in aluminium, although, it should be straightforward to generalize to the formation of stacking fault tetrahedra. Further, we will closely follow Kiritani's model (1973). This is to facilitate comparison with his computer results. For the same reason we will retain all the principal assumptions in his analysis. (Most of these can be justified. See Kiritani 1973 and also Ananthakrishna 1979a, b, c and d).

We write down the master equation for the problem of clustering. The rate constants are complicated functions of the sizes of the clusters and therefore it is not possible to follow the time dependence analytically. However, the stationary distribution can be calculated easily. This form still does not allow us to get either $\bar{N}$ or the average number of vacancies in the cluster $\langle n \rangle$, due to the fact that they are functions of the asymptotic value of single vacancy concentration N_1^{st} (which is in equilibrium with the remaining clusters). An attempt to calculate $\langle n \rangle$ formally leads to a differential equation whose solution gives $\bar{N}(N_1^{st}, T)$. If we assume that N_1^{st} very nearly follows Arrhenius plot (which should be true over a limited range of T) just as the equilibrium concentration of single vacancies, we can show that $\bar{N} \propto \exp - \bar{K}T$, above a temperature $T^* \simeq 210°K$. Both the value T^* and the qualitative nature of $\bar{N}$ agrees with experiments (Kiritani 1973). Since $\bar{N}$ and $\langle n \rangle$ are related through N_0, the quenched-in single vacancy concentration, the behaviour of $\langle n \rangle$ as a function of T can be calculated. The qualitative behaviour of N_1^{st} increasing as T is reduced and $\langle n \rangle$ decreasing as T is decreased below the plateau region also follow from our analysis.

2. The model

The model is in essence that of Kiritani which has two principal idealizations, namely, the absence of sinks and impurities, and the existence of only one mobile species. We shall also assume no loss of vacancies from the sample. These two idealizations make the problem transparent and mathematically tractable. For justification (or the conditions under which these assumptions are valid) we refer the reader to Kiritani (1973) and Ananthakrishna (1979a, b, c and d). We shall refer to the papers in this series as I, II, III and IV (Ananthakrishna 1979a, b, c, d respectively) and their respective equations by (I. n) etc. The notation used is the same as in I–IV. Following Kiritani (1973) we use the relation $n = 4\pi r^2/(a^2\sqrt{3})$, where a is the lattice parameter. The expression for the energy of formation of a vacancy loop we use is that of Schoeck and Tiller (1960) with n given by the above relation. Since, we will parametrize the constants involved, it is sufficient to write it in the form

$$\epsilon_n = An + Bn^{1/2} \ln cn, \tag{1}$$

where A, B and c are positive constants. The values of these constants we use are close to the actual values in the work of Schoeck and Tiller (1960). We shall assume that equation (1) holds for small n also. The binding energy b_n is given by (1.1).

$N_1, N_2, \ldots, N_n$ are the concentrations of 1–, 2–,..., n– unit clusters. These clusters evolve by absorbing and emitting single vacancy units. The rate equations are given by (I. 2–4). Since we do not have sinks, the total number of vacancies are conserved. This is represented by (I. 6). We stress that this result is independent of the form of the transition rates W's. In the case of formation of vacancy loops, we take the same choice of W's as that of Kiritani (1973)

$$W_{1n} = \nu \psi (n) Z_1 \exp (-E^M/KT), \tag{2}$$

and

$$W_{1n} = \nu \bar{\psi} (n) Z_2 \exp (-E^M + b_n)/KT), \tag{3}$$

where E^M is the migration energy of the mobile unit, ν is the attempt frequency, Z_1 and Z_2 are coordination numbers for hopping, and $\psi (n)$ and $\bar{\psi} (n)$ are the number of absorption and emission sites respectively. Following Kiritani (1973), we have taken the number of capture sites $\psi (n)$ to be the number of atomic sites on an equivalent circular loop whose radius is greater than that of the vacancy loop by two atomic units. $\bar{\psi} (n)$ is taken to be the number of atomic sites on the equivalent circular loop. Thus

$$\psi (n) = n + an^{1/2}, \; \bar{\psi} (n) = n. \tag{4}$$

Z_1 and Z_2 have been taken to be equal. In a realistic situation the number of absorption and emission sites are functions of the size of the cluster. For small n, these are expected to vary linearly with n. However, for large n the number of capture sites should be proportional to $n^{1/2}$. In spite of this, we will retain this assumption mainly because we would like to be able to compare our analytical results with

Kiritani's computer results. (Since we have used his model, we expect to find similar results for the same values of the parameters.) Clearly the method itself is not restrictive and the extension of the work to any form of ψ and $\bar{\psi}$ is straightforward including ψ and $\bar{\psi} \sim n^{1/2}$.

3. The stationary distribution

Consider (I.4) with the form of W's given above. We have modelled the process as a one-step Markoff process (Ananthakrishna 1979a–d). The stationary state is given by equating the right side of (I.4) to zero. This gives

$$(E^{-1} - 1)\, N_1^{st}\, W_{n+1\,1}\, N_n^{st} + (E - 1)\, W_{1n}\, N_n^{st} = 0,$$

or

$$(E - 1)\, [W_{1n}\, N_n^{st} - E^{-1}\, N_1^{st}\, W_{n+1\,1}\, N_n^{st}] = 0,$$

where E and E^{-1} are the forward and backward difference operators. Thus

$$W_{1n}\, N_n^{st} - E^{-1}\, N_1^{st}\, W_{n+1\,1}\, N_n^{st} = J. \tag{5}$$

This, then, represents the density flow from n to $n - 1$. Our problem is a ' one-step birth and death process ' with the transition probabilities from a state n' to n (of the master equation) given by

$$W(n/n') = r(n')\, \delta_{n,n'-1} + g(n')\, \delta_{n,n'+1}. \tag{6}$$

Here $g(n)$ and $r(n)$ correspond to the association and the dissociation rates (van Kampen 1976). Then

$$g(n) = \nu\, Z_1\, \psi\, (n + 1)\, N_1 \exp\, (-\, E^M/KT) = x_1\, N_1\, \psi\, (n + 1) \tag{7}$$

and

$$r(n) = \nu\, Z_2\, \bar{\psi}\, (n) \exp\, -\, (E^M + b_n)/KT = x_1 x_2\, (n)\, \bar{\psi}\, (n), \tag{8}$$

where

$$x_1 = \nu\, Z_1 \exp\, -\, E^M/KT \text{ and } x_2\, (n) = \exp\, -\, b_n/KT.$$

Equation (5) is not valid for $n = 1$ and 2. In this sense $n = 1$ is an artificial boundary. (However, if we impose (6) be true for $n = 1$, then we are forced to take $r(1) = 0$, since, it corresponds to dissociation and $N_{n=0} = 0$. Thus it should be classified as a natural boundary according to van Kampen (1976).) Generally artificial boundaries are relatively more difficult to solve. In the present case since the number of vacancies are conserved, we can assume that there is no probability leak at $n = 1$. Thus, we take $J = 0$ in (5). We shall later show that N_n^{st} thus determined does, in fact, satisfy the equation for $\dot{N}_1$.

Using (7) and (8) in (5) we get

$$N_n^{st} = \frac{\Pi_2^n g(i-1)}{\Pi_2^n r(i)} N_1^{st},$$

or
$$N_n^{st} = (N_1^{st})^n \left[\Pi_{i=2}^n (1 + \alpha i^{-1/2})\right] \left[\Pi_2^n x_2(j)\right]^{-1}. \tag{9}$$

Using (I.1)

$$\Pi_2^n x_2(j) = \exp\left[-\frac{nE_v^F}{KT} + \frac{\epsilon_n}{KT}\right],$$

where we have used $b_2 = 2E_v^F - E_{vv}$. (E_{vv} is the energy of formation of a cluster with two units.) Then

$$N_n^{st} = (N_1^{st})^n \left[\exp + (nE_v^F - \epsilon_n)/KT\right] \left[\Pi_2^n (1 + \alpha\, i^{-1/2})\right], \tag{10}$$

and is exact for any choice of ϵ_n. Using (1) and (10) we get

$$N_n^{st} = (N_1^{st})^n \left[\exp n\frac{E_v^F - A}{KT} - \frac{B}{KT} n^{1/2} \ln cn\right] \Pi_{i=2}^n (1 + \alpha i^{-1/2}). \tag{11}$$

It should be noted that N_n^{st} is independent of the energy of migration. This is not surprising since x_1 cancels off in the asymptotic rates. Only time scales over which clustering proceeds depend on x_1, (*i.e.* t can be scaled by x_1). Our earlier calculation (Ananthakrishna 1979a–d) also supports this result. We shall now show that N_n^{st} determined by (11) satisfies the equation for N_1. Using (9) in (I.2) we get

$$\sum_2^n W_{m1} N_1^{st} N_{m-1}^{st} = \sum_2^{n-1} (m + \alpha m^{1/2}) N_1^{st} (N_1^{st})^{m-1}$$

$$\Pi_{p=2}^{m-1} (1 + \alpha p^{-1/2}) \left[\Pi_2^{m-1} x_2(j)\right]^{-1}$$

$$= \sum_{m=2}^n m (N_1^{st})^m \Pi_{p=2}^m (1 + \alpha p^{-1/2}) (\Pi_{j=2}^{m-1} x_2(j))^{-1}.$$

This is equal to

$$\sum_{m=2}^n W_{1m} N_m^{st} = \sum_{m=2}^n m \Pi_{p=2}^m (1 + \alpha p^{-1/2}) (N_1^{st})^m (\Pi_2^m x_2(j))^{-1}$$

$$\times \exp -(E_v^F - \epsilon_n + \epsilon_{n-1})/KT,$$

$$= \sum_{m=2}^n m \Pi_2^m (1 + \alpha p^{-1/2}) (N_1^{st})^m (\Pi_{j=2}^{m-1} x_2(j))^{-1}.$$

Thus the choice of $J = 0$ in (5) is consistent with (I.2). The stationary distribution can be rewritten using continuum approximation when n is large. (This would be valid except for very low temperatures. Generally, $\langle n \rangle$ is always large enough to permit this approximation.)

Consider

$$\ln \prod_m^n (1+\alpha m^{-1/2}) = \Sigma \ln (1+\alpha m^{-1/2}) = \int \ln (1+\alpha m^{-1/2})\, dm.$$

From this, we get

$$\prod^n (1+\alpha m^{-1/2}) \simeq \bar{c}\, (1+\alpha n^{-1/2})^n \, (\exp \alpha n^{1/2}) \, (\alpha+n^{1/2})^{-\alpha^2},$$

where $\bar{c}$ is a constant whose value is determined at $n = 2$. For large n, $(1+\alpha n^{-1/2})^n \to \exp(\alpha n^{1/2})$. Thus

$$N_n^{st} = \bar{A}\, (\alpha+n^{1/2})^{-\alpha^2} \, (N_1^{st})^n \exp [bn + 2\alpha n^{1/2} - dn^{1/2} \ln cn], \tag{12}$$

where $b = (E_v^F - A)/KT$, $d = (B/KT)$, and $\bar{A}$ a normalization constant. In (12) N_1^{st} is not known and has to be determined self consistently. As it stands, the distribution *does not* have the feature that as $n \to \infty$, $N_n \to 0$. However, a correct self-consistent choice of N_1^{st} should ensure this. (At the least, as $n \to \bar{n}$, $N_n^{st} \to 0$ where $\bar{n}$ is the maximum of the physically allowed values of n). We shall assume that such an N_1^{st} can be found (at present), then we can assume that both $\bar{N} = \Sigma N_n$ and $\Sigma n N_n = \langle n \rangle \bar{N}$ exist. (From now on, $\langle n \rangle$ and $\bar{N}$ are used for stationary states.) Then

$$\bar{N} = \bar{A}\, \Sigma \frac{\exp [(b + \ln N_1)\, n]}{(\alpha + n^{1/2})^{\alpha^2}} + (2\alpha - d \ln c)\, n^{1/2} - dn^{1/2} \ln n. \tag{13}$$

Let $$\exp (b + \ln N_1) = z \tag{14}$$

and $$\exp (2\alpha - d \ln c) = x. \tag{15}$$

Any attempt to calculate $\langle n \rangle$, then leads us to an equation of constraint that $\bar{N}$ has to satisfy. This constraint is in the form of a differential equation for $\bar{N}$. Differentiating (13) we get

$$\langle n \rangle \bar{N} = \frac{\partial \bar{N}}{\partial \ln z} = \frac{\partial^2 \bar{N}}{(\partial \ln z)^2}. \tag{16}$$

This is a 'diffusion' equation in the variables $\ln x$ and $\ln z$. A solution of this equation is

$$\bar{N} = \frac{\bar{B} \exp - \dfrac{(\ln x)^2}{4 \ln z}}{(4 \ln z)^{1/2}} = \frac{\bar{B} \exp - \dfrac{\left(2\alpha - \dfrac{B \ln c}{KT}\right)^2}{4 (\ln N_1^{st} + (E_v^F - A)/KT)}}{\left[4 \left(\ln N_1^{st} + \dfrac{E_v^F - A}{KT}\right)\right]^{1/2}}, \tag{17}$$

$\overline{B}$ is a normalization constant to be determined either by boundary condition on $\overline{N}$ or by comparing with experimental data. Although the correct dependence of N_1^{st} on T would be needed, if $\overline{N}(T)$ is to be determined, in order to assess the qualitative feature of $\overline{N}(T)$, a qualitative behaviour of N_1^{st} is sufficient. To see this we assume that

$$\ln N_1^{st} = -\frac{E'}{KT}, \text{ then } \ln N_1 + \frac{E_v^F - A}{KT} = \frac{\overline{E}}{KT} \text{ with } \overline{E} > 0.$$

We should expect this from $N_1^{st}(T) > N_1^{st}(T)$, the equilibrium concentration of single vacancies. Then

$$\overline{N} \simeq \frac{\overline{B}(KT)^{1/2}}{2\overline{E}^{1/2}} \exp - \left(2\alpha - \frac{B \ln c}{KT}\right)^2 KT/4\overline{E}. \tag{18}$$

Clearly, this function has a maximum. For temperatures T much larger than $B \ln c/2 K\alpha$,

$$\overline{N} \sim \frac{\overline{B}(KT)^{1/2}}{2\overline{E}^{1/2}} \exp - \overline{K}T, \tag{19}$$

where $\overline{K}$ is at most a weak function T. For temperatures T much smaller than $B \ln c/2K\alpha$, we see that

$$\overline{N} \sim \frac{\overline{B}(KT)^{1/2}}{2\overline{E}^{1/2}} \exp - \overline{K}T^{-1} \tag{20}$$

The temperature at which T^* is maximum is given by

$$T^* = B \ln c/2K\alpha. \tag{21}$$

Following Kiritani (1973), we choose $n = 4\pi r^2/a^2 \sqrt{3}$ then $\alpha = 8\pi^{1/2}/3^{1/4}$. The value of $B \ln c$ we use is 0·37. Then the maximum occurs at 200°K. (The value used by Kiritani for $B \ln c = 0{\cdot}54$. We wish to point out that if we use the above relation to define T^* for $B \ln c = 0{\cdot}54$, we get $T^* = 270$°K. This is exactly the temperature at which his computer results show a maximum).

Clearly, the assumption that $\ln N_1^{st} + b$ with $\overline{E}$ independent of temperature cannot be expected to hold. Indeed, as the temperature is lowered we should expect that the fraction of single vacancies, which go to form clusters decreases, *i.e.* N_1^{st} should increase. As $T \to 0$, $N_1^{st} \to N_0$. (Note that we have no sinks). But at higher temperature N_1^{st} is small compared to N_0 and should not be expected to vary much over a limited range. In the following, we shall determine N_1^{st} in an approximate way using (I. 2). Using the form W_{1n} and W_{n1}, we have

$$\Sigma \psi(m) N_1^{st} N_{m-1}^{st} = \Sigma \overline{\psi}(m) N_m.$$

Consider the left side

$$\sum_2 (m+\alpha m^{1/2})\, N_1^{st}\, N_{m-1}^{st} = (2+\alpha 2^{1/2})\,(N_1^{st})^2 + \sum_2 [(m+1)+\alpha(m+1)^{1/2}] \times N_1^{st}\, N_m^{st},$$

or

$$(2+\alpha 2^{1/2})\,(N_1^{st})^2 + N_1^{st}\,\bar{N}\,\langle n\rangle + \alpha\,\langle n^{1/2}\rangle\, N_1^{st}\,\bar{N} \simeq N_1^{st}\,\bar{N}\,[\langle n\rangle + \alpha\,\langle n\rangle^{1/2}] + (N_1^{st})^2\,(2+\alpha 2^{1/2}). \tag{22}$$

Here we have made two approximations namely changing the summation index does not alter the averages, (since the upper limit of the summation is large) and that $\langle n^{1/2}\rangle = \langle n\rangle^{1/2}$. We shall drop $(N_1^{st})^2$ compared to N_1^{st}. The right side is

$$\bar{N}\,\langle n \exp - b_n/KT\rangle \simeq \bar{N}\,\langle n\rangle\,\langle \exp - b_n/KT\rangle. \tag{23}$$

We shall use ϵ_n given by (1) in (I. 1) and using $\epsilon_n - \epsilon_{n-1} \to \frac{d\,\epsilon_n}{dn}$, we get

$$b_n = E_v^F - A - \frac{B}{2n^{1/2}}[\ln cn + 2]. \tag{24}$$

This holds as long as n is large. (The same expression results in the limit of large n, if we use (1) and the approximation $(n-1)^{1/2} \simeq n^{1/2} - \frac{1}{2}\, n^{-1/2}$ and $\ln (1-1/n) \simeq (1/n)$. We have checked that we can use this form only when $n > 250$. However, it is possible to fit the following expression for b_n to that of b_n obtained when (1) and (I.1) are used. This holds down to $n = 10$.

$$b_n = -0{\cdot}185 + 0{\cdot}31\,(1 - \exp - (n/n_0)^{1/2}),\ n_0 = 30. \tag{25}$$

Table 1 gives the values calculated using (25) and (1). Thus, we wish to use (25) rather than (24). Using (25) in (23), and using the approximation of taking average inside the expression, we get after using (21)

$$N_1^{st} = \frac{\exp\left[-\dfrac{0{\cdot}495}{KT} + \dfrac{0{\cdot}31}{KT}\exp - \langle n\rangle^{1/2}/n_0^{1/2}\right]}{[1+\alpha\,\langle n\rangle^{-1/2}]}, \tag{26}$$

Table 1. Comparison of the binding energy calculated from equation (1) with the empirical relation given by equation (25).

n	b_n calculated from equations (1) and (I. 1)	b_n calculated from equation (25)
10	0·303	0·320
20	0·357	0·357
30	0·382	0·387
50	0·408	0·409
90	0·433	0·44
180	0·455	0·468

(We have used the value of $E_v^F = 0{\cdot}67$ eV). We can use equation (26) in (17) to obtain

$$\bar{N} = \frac{\bar{B}\exp - \left\{\left(2\alpha - \frac{B\ln c}{KT}\right)^2 \Big/ \left[4\left(\frac{0{\cdot}1}{KT} + \frac{0{\cdot}31}{KT}\exp - \frac{\langle n\rangle^{1/2}}{n_0^{1/2}}\right) - 4\ln\left(1+\alpha\langle n\rangle^{-1/2}\right)\right]\right\}}{2\left\{\left[\frac{0{\cdot}1}{KT} + \frac{0{\cdot}31}{KT}\exp - \frac{\langle n\rangle^{1/2}}{n_0^{1/2}}\right] - \ln\left(1+\alpha\langle n\rangle^{-1/2}\right)\right\}^{1/2}} \quad (27)$$

For high temperatures (around the room temperature) one can use the approximation $N_1 \ll N_0$, thus (I.6) implies $\langle n\rangle = N_0/\bar{N}$. (Note $f(t) = \langle n\rangle\,\bar{N}$ in (I.6)). In this approximation (27) is a transcendental equation whose solution can be easily obtained if the normalization constant can be fixed. Normally this can be determined using the boundary conditions. In the present case both $T=0$ and $T=T_Q$ pose problems. This formula in principle cannot hold for $T=T_Q$ since there is no supersaturation. At the point $T=0$, $\bar{N}=0$, again $\bar{B}$ cannot be determined. Thus, we have to use an arbitrary point to determine $\bar{B}$. ($\bar{B}$ should have dimensions of concentration.) We shall later discuss this point again. We have used the value $\bar{B} = 10^{-7}$ cm^{-3}, at $T=213$°K. A plot of $\bar{N}$ as a function of T is shown in figure 1. The nature of the curve qualitatively agrees with the experimental curve shown by the dashed curve. (The circles are the experimental points.) *Even* within this approximation, we find that for low temperatures, $\bar{N}$ drops sharply below 130°K (not shown in the figure). Although this trend should be expected, we cannot calculate $\langle n\rangle$ from $\bar{N}$ because of the breakdown of the approximation for low T. For low temperatures, $\langle n\rangle$ should also be expected to decrease which implies from relation (I.6) that $N_1^{st} = \langle n\rangle\,\bar{N} - N_0$, increases until it reaches a value of N_0 as $T\to 0$. This feature about $\langle n\rangle$ cannot be calculated. However, we will show

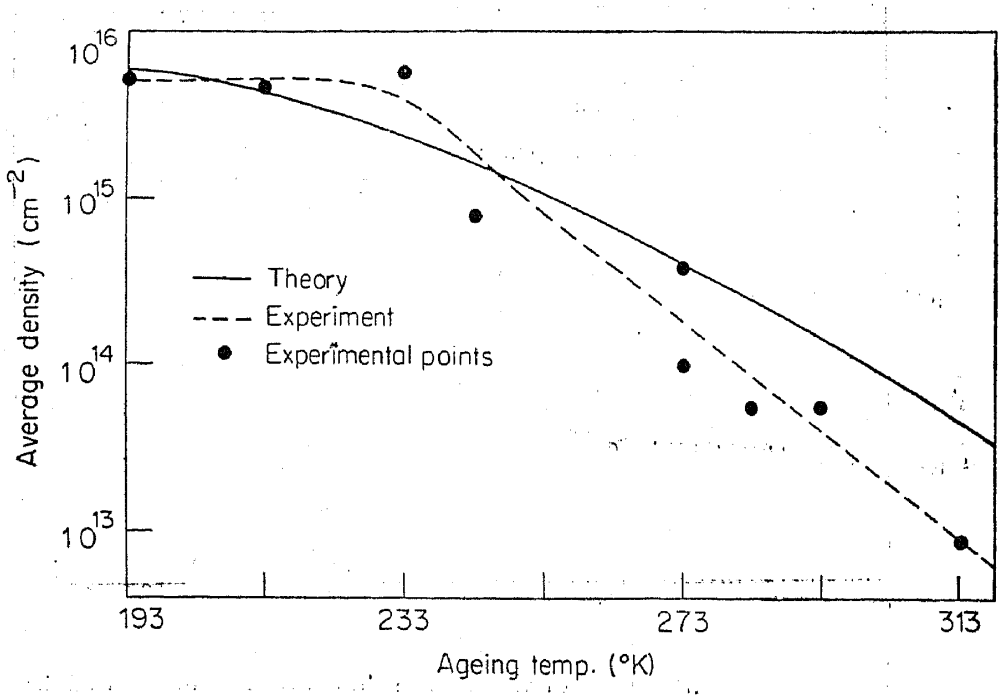

Figure 1. Density of vacancy loops as a function of the ageing temperature.

below that this qualitative feature can be obtained. For high temperature $N_1^{st} \ll N_0$ and thus $\langle n \rangle$ can be calculated from the relation $\langle n \rangle = N_0/\overline{N}$. A plot of this is shown in figure 2, along with the experimental curve (dashed line) and the experimental points.

4. Discussion

We show, in what follows, that it is possible to obtain the qualitative behaviour of $\langle n \rangle$. Using $\overline{N}$ from (17) and using (16) we get

$$-\frac{1}{2\,(\ln N_1^S + b)} + \frac{\left(2\alpha - \dfrac{B \ln c}{KT}\right)^2}{4\,(\ln N_1^{st} + b)^2} = \langle n \rangle.$$

Clearly, the first term is small. Thus

$$\ln N_1^{st} + b = \frac{\left|2\alpha - \dfrac{B \ln c}{KT}\right|}{2\,\langle n \rangle^{1/2}}. \tag{28}$$

Using equations (22) to (24), and using the approximation of taking the averages inside, we get

$$\ln N_1^{st} + b = \frac{B}{2KT\,\langle n \rangle^{1/2}} [\ln c\,\langle n \rangle - 2]. \tag{29}$$

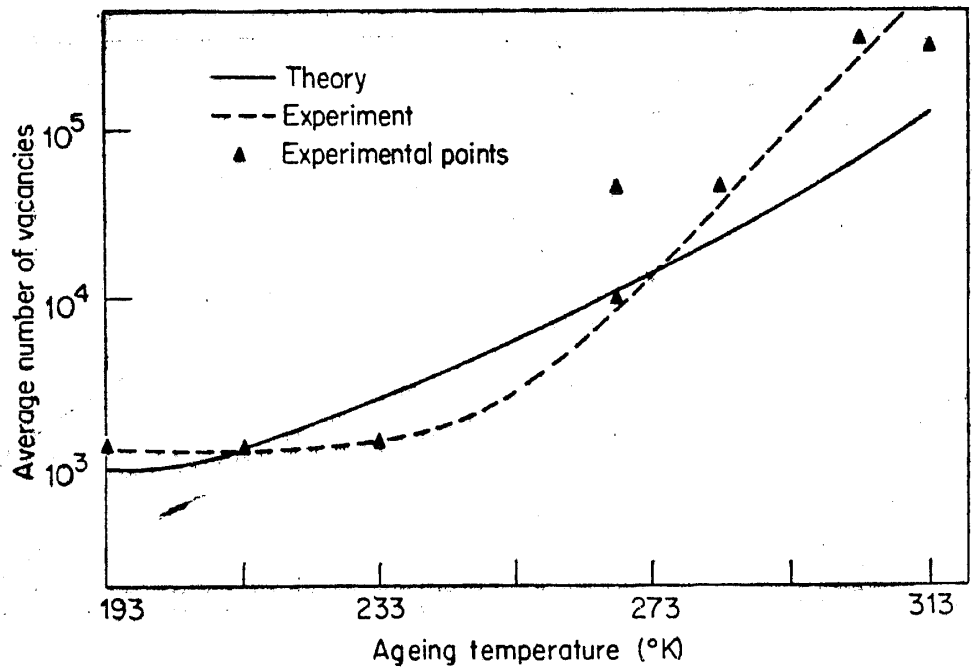

Figure 2. Average number of vacancies in the cluster as a function of the ageing temperature.

(Here we have ignored the factor $1/(1 + \alpha \langle n \rangle^{-1/2})$ in the expression for N_1^{st}). Equating (29) with (28), we get

$$\langle n \rangle = \exp\left[\frac{KT\left|2\alpha - \dfrac{B \ln c}{KT}\right|}{B} - 2 - \ln c\right]. \tag{30}$$

The qualitative nature of $\langle n \rangle$ predicted by this expression appears to be correct. For low temperatures ($T < T^*$) the term $B \ln c$ contributes a constant term and $2\alpha KT$ decreases steadily as T is decreased. For $T > T^*$, again $2\alpha KT$ increases steadily. Further, it also predicts a broad shoulder for $T = T^*$. Thus the qualitative behaviour is correct. However, as for the magnitude, there is a need for a scale factor of five multiplying the exponent. We believe that this is related to the inadequacy in determining $\overline{B}$ in the equation for $\overline{N}$ in (27). In principle, $\overline{B}$ should be a function of N_0 (T_Q). This can be seen from the fact that, if the quenching temperature is reduced, (for a fixed ageing temperature) $\overline{N}$, decreases. Since $\overline{B}$ should have the same dimensions as $\overline{N}$, perhaps $\overline{B} \propto N_0$, and the remaining factor 10^{-3} can be absorbed in the the exponential. This can give rise to the above scaling factor. It should be pointed out that (25) determines $\ln N_1^{st} + b$ self consistently. Hence (30) should represent the functional form of $\langle n \rangle$ correctly. This also means N_1^{st} does increase at $T \to 0$, as is physically desired. From the present analysis, *it is clear that the plateau in $\overline{N}$ as a function of T results from the fact that the number of absorption sites is larger than that for emission.* As far as the determination of the distribution, it appears that we need more (or better) constraints or a better way of estimating N_1^{st} than we have established here. Although, we have proved these results for the case of the formation of vacancy loops, the results should hold for stacking fault tetrahedra with minor changes. It should be possible to extend these considerations to the formation of voids.

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