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CONDITIONS FOR DISLOCATION LOOP PUNCHING BY HELIUM BUBBLES*

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Under continuous helium but insufficient vacancy supply, bubbles in metals grow by pressure-driven athermal processes such as metal interstitial emission and dislocaton loop punching. To discuss the energetic conditions for these processes, the formation and interaction energies of bubbles and interstitial type metal defects are analyzed. The only condition which has been considered to date is that the decrease in the free energy of a bubble associated with the emission of an interstitial type metal defect must be equal to or larger than the formation free energy of the latter defect. Consideration of the elastic interaction energies results in additional conditions controlling loop punching for bubbles with radii larger than about 10 Burgers vectors. Possible modes for the loop punching process are discussed.

1. INTRODUCTION

Helium has been theoretically predicted[1] and experimentally confirmed[2] to be practically insoluble in metals. Therefore, it strongly tends to precipitate into helium-vacancy clusters and bubbles when it is introduced by direct injection or by (n,α) reactions. The energetics and formation kinetics of this process form the basis for the understanding of helium effects such as swelling, intergranular embrittlement, and blistering.

The character of the precipitation kinetics of helium in metals depends distinctly on the helium production and displacement rates as well as on temperature. For instance, at high helium production rates and/or low temperatures, diffusion-controlled clustering of thermal- or radiation-induced vacancies is too slow to contribute to the formation of helium bubbles. In this case, helium precipitation is associated with athermal processes such as spontaneous formation of Frenkel pairs, metal interstitial ejection,[3,4] and dislocation loop punching.[5,6] On the other hand, at low helium production rates and high temperatures, vacancy absorption is fast enough to prevent such athermal processes. In this case, the pressure within a growing bubble is kept close to the thermal equilibrium value.

A necessary condition for the emission of an interstitial type metal defect by a helium vacancy cluster or helium bubble is that the decrease of the free energy of the helium vacancy cluster or helium bubble associated with such a process must at least be equal to (or larger than) the formation free energy of the metal interstitial defect. This condition has been discussed in detail recently for loop punching by small bubbles down to sizes of about 1 nm in diameter.[7]

On the other hand, the formation of an interstitial type metal defect has to occur at the bubble surface and this defect has eventually to overcome an energetic barrier to become separated from the bubble. In fact, computer simulation studies show that helium clustering results in strongly bound complexes of helium-vacancy and metal interstitial

*Research sponsored by the Division of Materials Sciences, U.S. Department of Energy, under contract W-7405-eng-26 with the Union Carbide Corporation and by the Office of Fusion Energy under contract DE-AC02-82ER50282 with the University of Wisconsin.

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clusters.[3] To date it has not been clarified whether and how such complexes finally grow and transform into stages from which they can dissociate completely.

In the present paper, first the necessary energetic conditions for the separation of interstitial type metal defects from overpressurized helium bubbles will be briefly reviewed. Then the energetic conditions for the formation of close complexes consisting of a helium bubble and metal interstitial clusters or dislocation loops will be set up. Finally, possible modes for complete dissociation will be sketched. Since elastic continuum theory will be used throughout the discussion, the resulting conditions cannot be taken to be fully quantitative but may be expected to describe the functional dependencies correctly.

2. FORMATION ENERGIES OF HELIUM BUBBLES AND METAL INTERSTITIAL DEFECTS

Consider the dissociation of a bubble of volume $V_B = 4\pi\, r_B^3/3$ into a bubble increased by the volume of N metal atoms, $N\Omega$, and a defect cluster consisting of N metal interstitials. A necessary condition for this process to occur is that the change in the free energy associated with it is zero or negative,

$$\Delta F_\infty = F_B(V_B + N\Omega) + F_D(N\Omega) - F_B(V_B) \leq 0 \qquad (1)$$

where F_B and F_D are the formation free energies of bubbles and metal interstitial clusters, respectively.

If the bubble is sufficiently large, the bubble formation free energy F_B is given by the contributions of helium inside the bubble volume, F_{He}, and the metal surface confining it, $F_S = \gamma$, where γ is the specific surface-free energy of the metal. Using this, Eq. (1) results in a condition for the average $\langle p \rangle$ of the helium pressures in the initial and final states[7]

$$\tilde{p} \equiv \langle p - 2\gamma/r_B \rangle \geq F_D/N\Omega \qquad (2)$$

For bubbles with radii r_B below 5 nm, curvature corrections to the bubble metal interface free energy become increasingly important.[7] We neglect them in the following to focus interest to the most important points of the problem under consideration.

To complete the condition expressed by Eq. (2), the number N of metal interstitials within the cluster and its formation free energy F_D must be specified. For a single interstitial, N = 1, the formation free energy is about $\mu\Omega/2$ where μ is the shear modulus of the metal resulting in

$$\tilde{p}_I \gtrsim \mu/2 \qquad (3)$$

as a condition for metal interstitial emission.

Due to an increasing binding of interstitials to their clusters, $F_D/N\Omega$ decreases with increasing N thus reducing the pressure necessary for interstitial cluster ejection. It has been shown[8] that in fcc metals small clusters form three-dimensional structures whereas larger clusters tend to assume two-dimensional platelet-like structures defining the transition to dislocation loops which are energetically most favorable for large N.

The formation energy of a dislocation loop, F_L, consists of an elastic energy, a dislocation core energy, and eventually a stacking fault energy neglected in the following. The two former contributions may be summarized in one unique expression which for circular edge dislocation loops of Burgers vector $\underline{b}$ and radius r_L containing $N = \pi b r_L^2/\Omega$ interstitials takes the form

$$F_L = \frac{\mu b^2 r_L}{2(1-\nu)} \ln \frac{r_L}{r_0} \qquad (4)$$

where ν=Poisson's ratio of the metal and r_0 is an effective core radius. With

$r_0 \approx 0.1$ b, this equation describes the formation energy of two-dimensional metal interstitial clusters down to sizes where the elastic continuum theory breaks down. The rough but simple approximation $F_L \approx \pi\mu b^2 r_L$ to Eq. (4) applicable in the range $4 < r_L/b < 20$ results in[7]

$$\tilde{p}_L \gtrsim \mu b / r_L \qquad (5)$$

as a condition for loop punching. Accordingly, the threshold pressure p_L for this process should decrease with increasing loop radius r_L. However, since such a loop originates in the bubble, its radius cannot be much larger than that of the bubble $r_L \approx r_B$. Additional conditions result from consideration of interaction energies between bubbles and metal interstitial defects as will be shown in the following.

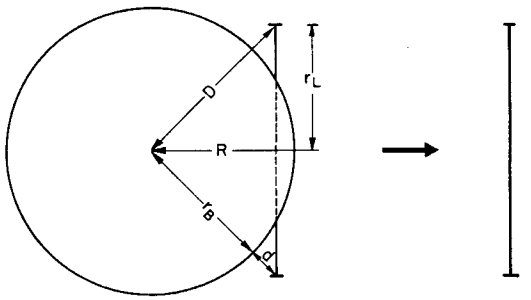

FIGURE 1
Scheme of the loop punching mechanism: formation and expulsion.

3. INTERACTION ENERGIES OF HELIUM BUBBLES AND METAL INTERSTITIAL DEFECTS

The energy of a complex of a helium bubble and a metal interstitial defect can be well described in terms of the formation and interaction energies of the constituents. The interaction energy is of elastic nature and consists of contributions due to the permanent strain fields characterizing the isolated defects, $E_{BD}^{(1)}$, called permanent or first order elastic interactions, and contributions due to additional strain fields around defects elastically polarized by the strain fields of the other defects, called induced, modulus or second order elastic interactions.[9,10] A bubble is elastically polarized by the strain field of a metal interstitial defect and, vice versa, inducing interaction energies $E_{BD}^{(2)}$ and $E_{DB}^{(2)}$, respectively. An elastic interaction of a defect with a boundary such as the bubble surface is also often referred to as an elastic image interaction[9], $E_{BD}^{(2)} \rightarrow E_{BD}^{Im}$.

The change in free energy associated with the formation of complexes of a helium bubble and a defect cluster consisting of N metal interstitials may then be written as

$$\Delta F = \Delta F_B + F_D\,(N\Omega) + E_{BD}^{(1)} + E_{BD}^{Im} + E_{DB}^{(2)} \qquad (6)$$

with $\Delta F_B = -\tilde{p} N\Omega$.

To specify the three interaction energy terms at the right hand side of this expression, the strain fields around bubbles and metal interstitial defects must be known. Spherical bubbles and isotropic metal interstitial defects represent centers of dilation which in isotropic media are characterized by displacement fields of the form[10]

$$\underline{u} = \frac{\hat{p}}{4\mu} \frac{r_B^3/D}{R^3} \underline{R} \qquad (7)$$

where $\hat{p}$ is the pressure transferred to the metal matrix adjacent to the defect and $\underline{R}$ is the position vector measured from its center. For a bubble, $\hat{p} = p - 2\hat{\gamma}/r_B$, where $\hat{\gamma}$ is the surface tension of the metal. Since $\hat{\gamma}$ is not known, we set $\hat{\gamma} = \gamma$ and $\hat{p} = \tilde{p}$. The strain field obtained by differentiating Eq. (7) is of pure shear character.[9] As a consequence the first order interaction between such dilation centers is zero, and the interaction energy is completely governed by second order interactions.

The image interaction of an isotropic defect with a bubble is attractive everywhere. It reaches $E_{BD}^{Im} = -E_D/3$ for $\nu = 1/3$ when touching the bubble surface[9] if the defect radius is small compared with the bubble radius, $r_D \ll r_B$. Single dumb-bell interstitials and small interstitial clusters in fcc metals have been shown to have a high elastic shear polarizability α_S[11] resulting in attractive interaction energies $(-\alpha_S \varepsilon_S^2)$ with the shear strain fields ε_S of other interstitial defects[12] or over-pressurized bubbles. Using Eq. (7) we can estimate the change in the free energy associated with the formation of a single interstitial or an isotropic interstitial cluster bound to the surface of a bubble to be

$$\Delta F \approx -\tilde{p}N\Omega + 2E_D/3 - 3\alpha_S\tilde{p}^2/(8\mu^2) \qquad (8)$$

with $E_D \approx \mu\Omega/2$ and α_S between 15 $\mu\Omega$ and 20 $\mu\Omega$[11] $\Delta F \lesssim 0$ results in $\tilde{p} \gtrsim \mu/6$.

In clustering the energy per single interstitial, E_D/N, is reduced due to their polarization induced interaction[12] but on the other hand the elastic polarizability decreases with increasing N. Accordingly, the binding of an interstitial to its cluster is weaker close to than far off an overpressurized bubble. Taking 1/3 of the formation energy of a single interstitial for the energy per interstitial in a large cluster[8] and neglecting its elastic polarizability we obtain from $\Delta F < 0$ the condition $\tilde{p} \gtrsim \mu/9$.

Single interstitials or three-dimensional interstitial clusters formed at the bubble surface will stay there due to the attractive elastic second order interaction. This is in agreement with observations made in computer simulation studies.[3]

The strain field of a circular dislocation loop is described by elliptical integrals[13] and becomes simple only close to, far from or on the axis of the loop. It is, however, not needed explicitly to calculate the first order interaction energy between a bubble and a loop, $E_{BL}^{(1)}$, since this can be represented by[13]

$$E_{BL}^{(1)} = -\int \underline{\underline{\varepsilon}}_B \, d\underline{\underline{P}} \qquad (9)$$

where $\underline{\underline{\varepsilon}}_B$ is the strain field of the bubble and $dP_{ij} = \sum_{kl} C_{ijkl} b_k dA_l$ is the dipole force tensor of an infinitestimal dislocation loop of Burgers vector $\underline{b}$ and surface element $d\underline{A}$ in a medium with elastic constants C_{ijkl}. For a circular edge dislocation loop in an isotropic medium integration of Eq. (9) yields a repulsive interaction which can be combined with ΔF_B in the expression $-\pi b r_L^2 (1 - r_B^3/D^3)\tilde{p}$ where D is the distance between the dislocation core and the bubble center (Fig. 1).

To determine the image interaction, E_{BL}^{Im}, the image field of a loop in the presence of a bubble surface must first be calculated. The procedure is analogous to that developed for the interaction of a loop with a void.[14] Since it is complicated in the general case, details will be presented elsewhere.[15] For the present discussion only the most important features are needed. For instance, the image force is attractive everywhere; it is dominant close to the bubble surface whereas the elastic first-order interaction force is dominant far from the bubble. If the distance of the dislocation line from the bubble surface, $d = D - r_B$, is small compared with r_B and r_L, the problem reduces to that of the image interaction of a straight dislocation with a plane surface.[13] Combining in this case E_L and E_{BL}^{Im} and neglecting the elastic polarizability of loops, $E_{LB}^{(2)} = 0$, we obtain for the free energy change on forming a loop close to the bubble surface

$$\Delta F \approx -\pi b r_L^2 (1 - r_B^3/D^3)\,\tilde{p} + \frac{\mu b^2 r_L}{2(1-\nu)} \ln \frac{d}{r_0} \qquad (10)$$

where the core radius r_0 of the loop is estimated to range from 0.1 b to 0.5 b for small and large loops, respectively. With $d \approx b$, $\Delta F \lesssim 0$ yields an approximate condition for the spontaneous formation of a loop adjacent to the bubble surface

$$\tilde{p}_L \gtrsim \frac{\mu}{6\pi(1-\nu)} \frac{r_B}{r_L} \ln \frac{b}{r_0} \sim (0.05 \text{ to } 0.2)\, \mu \frac{r_B}{r_L} \quad (11)$$

Accordingly, the pressure required decreases with increasing loop radius and becomes lowest for $r_L \approx r_B$. The minimum value is about equal to the threshold pressures for the formation of metal interstitials and clusters of them close to the bubble surface, i.e. about 1/2 the theoretical shear strength, $\mu/2\pi$, indicating a transition of the metal matrix adjacent to the bubble into a state of high crystal disorder.

Generally, a loop formed at the bubble surface in accordance with Eq. (11) will stay there since the force between both is attractive near the bubble. This is illustrated in Fig. 2 which shows the geometry of bubble-loop configurations obeying $\Delta F = 0$ for $r_0 = 0.5b$, $r_B = 50b$, and various values of $\tilde{p}$. Accordingly, a loop for which $\Delta F = 0$ at the bubble surface ($d/r_B = b/r_B = 0.02$) is repelled from the bubble only above a certain, though relatively small distance from the bubble surface ($d/r_B \approx 0.05$). The pressure required to form a loop at the position of zero force (maximum of the curves) is about twice that required to form it at $d = b$.

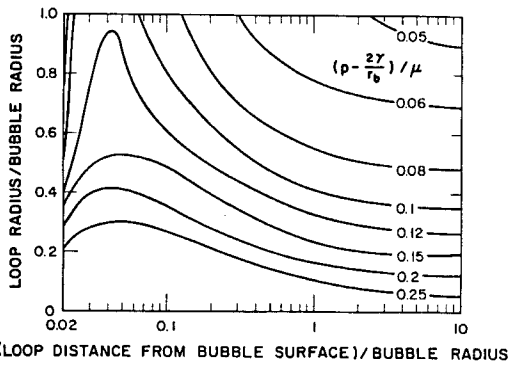

FIGURE 2
Geometry of bubble-loop configurations which can be formed without change in free energy, $\Delta F = 0$, for $r_0 = 0.5b$, $r_b = 50b$, and various pressures. For a given pressure the change in free energy is positive below and negative above the corresponding line, respectively.

4. CONDITIONS FOR THE EXPULSION OF A LOOP FROM A BUBBLE

A condition for complete dissociation of a loop from a bubble is that the force acting between them must be repulsive over the whole dissociation path (i.e., $\partial\Delta F/\partial R \lesssim 0$). Since the attractive image force counteracting the repulsive first order force increases with decreasing distance between the loop and the bubble surface it appears to be most important to fulfill this condition for the smallest possible distance, $d = b$. Using Eq. (10) we derive from $\partial\Delta F/\partial R \lesssim 0$ and $\partial^2\Delta F/\partial R^2 \lesssim 0$ for $d = b$

$$\tilde{p}_L \geq \frac{\mu}{6\pi(1-\nu)} \frac{r_B}{r_L} \text{ for } r_B/b \geq 6 \,. \quad (12)$$

The first condition is similar to the above one, Eq. (11), but is probably more reliable since the uncertain core radius r_0 has canceled. As for the formation of a loop, the lowest pressure for its expulsion is required for $r_L \approx r_B$, namely $\tilde{p} \approx \mu/4\pi$.

It depends upon the value of r_0 whether the threshold pressure for the formation of a loop is lower or higher than for its expulsion. In the first case a loop can be expelled only after the formation of one or more additional loops close to the bubble associated with an increase in the pressure up to values fulfilling the conditions (12).

For small bubbles with single dislocation loops, $\partial\Delta F/\partial R \lesssim 0$ cannot be fulfilled as indicated by the second part of the conditions (12). In this case a pile-up of loops around the bubble is required to establish a repulsive force for the expulsion of the most outer loop. This process is controlled by the condition for the formation of a completely dissociated loop as given by Eq. (5).

Combining the conditions, Eqs. (5) and (12), we are able to sketch the threshold pressure for loop punching as a function of the

bubble radius. This is done in Fig. 3. For small bubbles the threshold pressure decreases with increasing bubble size due to the decrease in the loop formation energy per interstitial for increasing loop size. For large bubbles, $r_B \gtrsim 10b$, an approximate constant threshold pressure is required for the expulsion of loops.

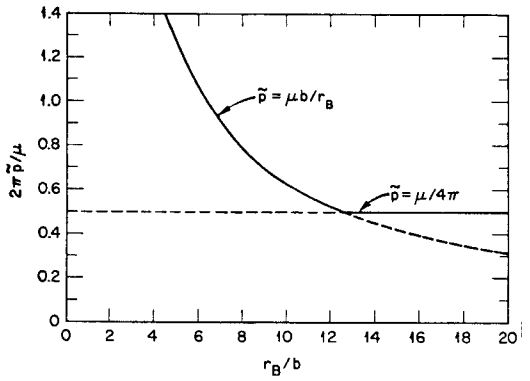

FIGURE 3
Threshold pressures for loop punching, $\tilde{p} = (p - 2\gamma/r_B)$, versus bubble radius, r_B, in units of the theoretical shear strength, $\mu/2\pi$, and the Burgers vector, b, respectively (schematically).

The details of the loop formation process close to the bubble surface are not yet fully understood. For small bubble sizes, however, it seems to be clear that this process starts with the formation of single interstitials and their agglomeration into small three-dimensional clusters staying near the bubble surface. Beyond a certain number of interstitials per cluster (say enough to form an equatorial loop) it is energetically more favorable to transform the three-dimensional cluster into a loop which will be expelled if a critical pressure in the bubble is reached. For large bubbles it is not clear whether this pre-stage of a three-dimensional interstitial cluster is required for loop formation.

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