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LETTER TO THE EDITORS – LETTRE AUX REDACTEURS

THE SHAPE OF AN OVERPRESSURIZED BUBBLE

There are many circumstances in which radiation-induced gas bubbles may contain an excess gas pressure. Such bubbles may be created by migration and coalescence at low temperatures [1] or by irradiation at high gas/damage ratios. They are important because they may lead to blistering [2], breakaway swelling [3] or dislocation loop punching [4]. The overpressure may persist almost indefinitely in the temperature range (below $0.5T_m$) where few vacancies are available but surface diffusion is rapid enough to permit bubble migration [5]. The shape of such bubbles is important because it may determine the rate-controlling process for bubble migration: A faceted bubble whose migration rate is limited by ledge nucleation on its faces may have a mobility several orders of magnitude less than an equivalent spherical bubble [5].

A simple consideration of the effect of a gas overpressure leads to the following argument: at equilibrium the gas pressure, p, balances the "surface tension forces", Γ, and no elastic strain need be stored in the lattice around the bubble. For a spherical bubble at a high temperature where the surface stress (tension) is equal to the surface energy, γ, then $\Gamma = 2\gamma/r$, or at equilibrium $p = 2\gamma/r$. More generally, for a faceted bubble with i faces of area A_i, $\Gamma = \Sigma_i\, \gamma_i A_i$. If, at a lower temperature, $p > \Gamma$ then elastic strain energy will be stored in the lattice and to minimize the stored energy we should minimize $(p - \Gamma)$. The bubble can achieve this without growing by maximizing Γ, i.e. by developing high-energy facets (e.g. by becoming spherical) or by developing large area faces (e.g. by becoming irregular). This argument (briefly: overpressurised bubbles tend to become sperhical) has been presented more rigorously by Nelson et al. [6]. It is the purpose of this note to point out that it is not the elastic stored energy which must be minimised to determine the bubble shape, but the total free energy of the system, which includes the gas. From this point of view it can be seen that an overpressurised bubble will tend to become faceted rather than spherical.

There are four components which contribute to the total free energy, E, of a system containing one bubble. These are the free energy of the gas, F, the internal energy of the matrix without the bubble, F^1, the elastic energy stored around the bubble, Q, and the free energy of the bubble surfaces, Γ. Thus,

$$E = F^1 + F + Q + \Gamma\ .$$

Now consider a bubble of volume V_0 with sufficient gas to balance the surface stresses, at a sufficiently low temperature that it cannot change its volume by accepting vacancies. For this system F^1 is essentially constant and $Q = 0$. Let us now introduce more gas until the pressure is p, and then let the bubble expand elastically. As the elastic expansion occurs the gas free energy will drop, while the stored energy and surface energy terms will increase. We will then have

$$E + \Delta E = \text{constant} + F + \Delta F + \Delta Q + \Gamma + \Delta\Gamma\ . \qquad (1)$$

Now F is the free energy of the gas at pressure p and volume V_0, and hence $F = pV_0$. The surface energy term, Γ, is dependent on shape: Let us consider a bubble of volume $V_0 = a^3$ and surface area $A = 6a^2s$. Then for a cube the shape parameter s is unity, whereas for a sphere $s < 1$ and for a less regular shape $s > 1$. We can now write, for a bubble faceted on only one type of plane,

$$\Gamma = 6a^2 s\gamma = 6sV_0\gamma/a\ .$$

This ignores the well-established fact that surface energy, γ, is not necessarily equivalent to surface stress, σ, at low temperatures [7,8]. However since we have virtually no quantitative information about the relative magnitudes of γ and σ and since the simplification does not alter our conclusion, it is made here. The implications of considering both γ and σ are considered in more detail elsewhere [9].

The changes in energy ΔF, ΔQ, and $\Delta\Gamma$ as the pore expands can be determined by considering the work which the gas does as it expands against both the elastic deformation of the matrix and the surface tension of the bubble surfaces. Now if $\Delta V/V_0 \ll 1$

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