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State of a pressurized helium bubble in iron

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Abstract – We study the state of a nanometric helium bubble in bcc-iron as a function of temperature and He content using atomistic calculations. It appears that up to moderate temperatures the Fe lattice can confine He to solid state, in good agreement with known solid-liquid transition diagram of pure He. However, He in the bubble forms an amorphous phase, while an fcc structure is expected at the same temperature and He density. In addition, the He bubble forms a polyhedron whose morphology depends on either the surface energy or the elastic-plastic properties of Fe at either low or high pressure, respectively. Indeed, at high He contents the bubble surface breaks down at the mechanical stability limit of the Fe crystal, leading to a pressure decrease in the bubble.

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Helium is a substance with weak van der Waals attractive forces, which dictate its liquid and solid states. Solid He has an exceptionally high compressibility, giving rise to a wide range of densities [1]. Following experiments [2–4] and simulations [5–7], solid He has been found to exist in three different stable crystallographic allotropies, namely hexagonal close-packed (hcp), face-centered cubic (fcc), and body-centered cubic (bcc) depending on temperature and pressure. However, the behavior of He may differ when embedded in materials. Helium can be implanted or produced in metals as a result of transmutation reactions due to, *e.g.*, high-energy neutron irradiation. High He concentrations can lead to nanometric bubbles, which degrade the mechanical properties of the material [8,9]. This occurs in irradiation facing materials such as the one foreseen for the future fusion reactors, whose development is at its prime. Hence, the understanding of the behavior of He in materials is of paramount importance. At present, the first candidate material for irradiation facing components of fusion reactors is ferritic steel.

Previous computer simulations based on molecular dynamics [10,11] (MD) and *ab initio* [12] methods indicate that helium atoms in Fe attract each other and form clusters of a few atoms, which can then grow and coalesce to nanometric bubbles. Although the increase of the helium atomic density, often denoted by the

helium to vacancy ratio, decreases the binding energy of helium atoms in the cluster, still for ratios up to 6 the binding energy can be as high as 2.0 to 2.5 eV [10–12]. The increase in the He density leads to a pressurized bubble, with subsequent elastic and plastic deformations in the surrounding lattice that may impact the passage of dislocations, vector of plasticity [13]. There is however a lack of knowledge on the state of the He bubble.

Here we study the phase state and morphology of a pressurized nanometric He bubble in bcc-Fe using MD simulations. Our sample consists of an Fe cubic simulation box of 70 bcc unit cells a side containing initially a He bubble, which is 2 nm in diameter, consistent with experimental observations in irradiated ferritic steels [8]. One should note that the way the He bubble is created in the simulation box differs from its natural formation in metals, which is due to atomic diffusion, nucleation and growth mechanisms. A range of He contents (1He:1v, 2He:1v, 3He:1v, 4He:1v, 5He:1v), quoted as the ratio of He atoms per vacancy in the bubble, and temperatures (10, 100, 200, 300, 700 K) are investigated. The samples are examined after an initial annealing at 700 K for 2 ps followed by a cooling down to the desired temperature using MD. When indicated a conjugate gradient relaxation was performed. We selected the many-body potentials derived by Ackland *et al.* [14] for the Fe-Fe interactions, and the pair potentials derived by Wilson-Johnson [15] and Beck [16] for Fe-He and He-He interactions,

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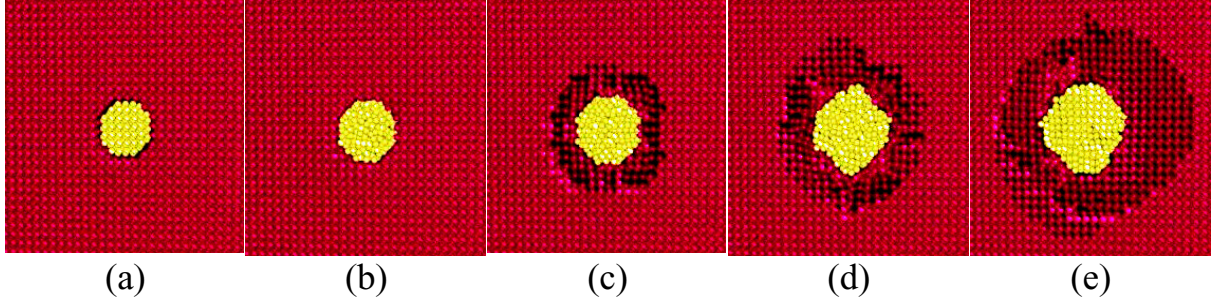


Fig. 1: (Colour on-line) Cross section of the 2 nm He bubble and surrounding Fe lattice after relaxation for various He contents; a) 1He:1v, b) 2He:1v, c) 3He:1v, d) 4He:1v, e) 5He:1v. Light (yellow) and dark (red) points represent He and Fe atoms, respectively.

respectively, as suggested by Morishita and collaborators [10]. Concerning the potential selection, as the Fe-He interface plays an important role in our investigations, some of the results are repeated with a newly developed Fe-He potential, Juslin-Nordlund [17], which gives a formation energy for He substitutional and He interstitial in Fe in closer agreement with *ab initio* calculations. Note that hereinafter all the results and discussions are related to the Wilson-Johnson potential unless Juslin-Nordlund potential is mentioned.

A view of the atomic structure of He in the bubble and surrounding Fe lattice after relaxation is given in fig. 1. In the case of the 1He:1v bubble (fig. 1a) a small gap between He and Fe atoms appears, which is due to the repulsion of He atoms by Fe. In other words, the pressure in the 1He:1v bubble does not overcome the repulsion by the surrounding Fe atoms. Note that it relates to the insolubility of He in metals [18]. With increasing He ratio this gap disappears (fig. 1b) and at higher He ratios, from 3He:1v (fig. 1c), a distortion of the Fe lattice surrounding the bubble is observed. There is a subsequent increase of the bubble volume with increasing He content, as clearly seen in figs. 1d and e, corresponding to 4He:1v and to 5He:1v bubbles, respectively.

The hydrostatic pressure on a given He atom is calculated according to $P_a = E_a/V_a$, where E_a and V_a are the total atomic energy and the atomic volume, respectively. The atomic volume is obtained by dividing the bubble volume by the number of He atoms in the bubble. The bubble pressure is then obtained by averaging the pressure over all He atoms. Only inner He atoms of the bubble are considered to avoid the influence of neighboring Fe atoms on the outer He atoms. The bubble pressure and atomic density as a function of He content and temperature are given in fig. 2. A sharp increase in the bubble pressure and density is observed for an increasing He content from 1He:1v to 2He:1v, whereas beyond this content the raise of pressure and density diminishes and further decreases, with a peak in pressure and density around 3He:1v. This relates to a visible failure of the surface of the bubble, which expands in the surrounding Fe lattice beyond 3He:1v. Iron thus deforms elastically

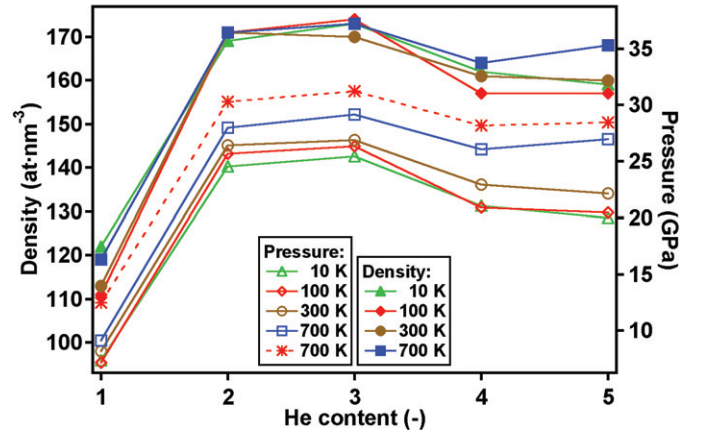


Fig. 2: (Colour on-line) The atomic density and pressure in the 2 nm bubble in Fe as a function of He content at different temperatures. Star marks: Juslin-Nordlund potential for Fe-He interaction.

and further plastically, partially releasing the bubble pressure and density. The maximum observed bubble pressure is 29 GPa, which relates to the 3He:1v bubble at 700 K. When the bubble surface breaks down, it is observed that He atoms penetrate into the surrounding Fe in mainly $\langle 100 \rangle$ directions (fig. 1d), and promoting self-interstitial atoms formation in the Fe lattice already initiated by the high pressure. The latter can lead to the formation of dislocation segments, a mechanism regarded as loop punching [19–21]. From 4He:1v to 5He:1v only a minor change in the bubble pressure and density is observed while its volume increases (fig. 1e). The pressure obtained using Juslin-Nordlund potential is at most 10% higher, except at 1He:1v, where it can be 30% higher at most, for the higher temperature. This may relate to the fact that Juslin-Nordlund potential induces a stronger repulsion of He atoms by Fe, which is noticeable only at low He contents. The fact that He can more easily penetrate Fe at high He content does not impact pressure.

The observed peak in pressure and density at 3He:1v relates to the mechanical stability limit of perfect bcc-iron under stress. The pressure at which the bubble surface

breaks can be estimated by [22]

$$p = (2\gamma + \mu b)/r, \quad (1)$$

where γ and μ are the free surface energy and the shear modulus of Fe, respectively, b is Burgers' vector of dislocation segments punched out from the bubble and r is the bubble radius. For the 3He:1v bubble, $r = 1.12$ nm, $b = 0.248$ nm, $\mu = 72$ GPa, obtained by MD calculation and $\gamma \approx 2.4$ J·m⁻², which is an average over various crystallographic surface orientations [23–25]. Note that the radius of 1.12 nm for 3He:1v bubble is observed after MD relaxation. Equation (1) gives 20.3 GPa for the mechanical stability limit. It should be noted that this value is few tens of percentage lower than the correct value for small bubble sizes due to the assumptions used to derive eq. (1) in ref. [22]. Indeed, this value should be compared with the pressures obtained by MD at finite temperatures, *e.g.*, 10 K, as eq. (1) was derived using elasticity theory without involving temperature. There is thus a fair match between the mechanical stability limit obtained by elasticity theory, eq. (1), and the MD simulation results, reported in fig. 2. Also, the comparison between two different potentials for Fe-He interaction shows that although the Wilson-Johnson potential does not reproduce the most recent *ab initio* characteristics so well as Juslin-Nordlund, both potentials give rise to the same conclusions in the estimation of the bubble pressure and of the mechanical stability limit.

With increasing temperature the He bubble can be expected to expand, leading to a decrease in the atomic density [26,27]. However, while our results show indeed that the atomic density of the bubble tends to increase, some exceptions are observed at low temperatures. This is due to the dominant thermal expansion of surrounding Fe lattice relative to the one of He bubble, which forces He to contract. The exceptions to this trend stem from various origins. In the case of 1He:1v, the gap that is seen in the Fe-He interface at low temperatures disappears at 100 K due to the bubble thermal expansion, causing a decrease in the atomic density. The 2He:1v bubble gives the lowest density change with temperature, owing to the equilibrium between the bubble pressure and the compression from the surrounding bcc lattice. Finally, beyond 3He:1v the bubble surface breaks, leading to a drop in atomic density at low temperatures. Besides He density, its pressure shows a consistent increase with increasing temperature for all He contents.

Self-diffusion coefficient was calculated using the mean square displacement (MSD), which is used to track the displacement of atoms. The MSD-time gradient gives the self-diffusion coefficient of atoms according to the following relation:

$$MSD = 6D_s t + C, \quad (2)$$

where D_s is the self-diffusion coefficient, t is the time and C is a fit constant. Helium self-diffusion coefficients, which

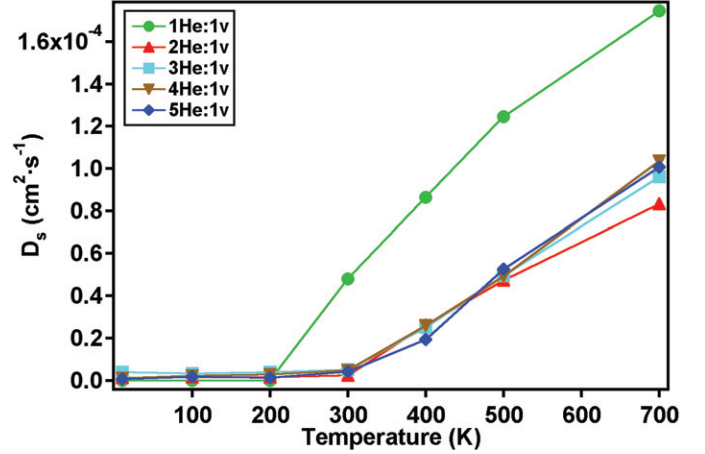


Fig. 3: (Colour on-line) He self-diffusion coefficients obtained from mean square displacement calculation for various He contents in the 2 nm bubble in Fe as a function of temperature.

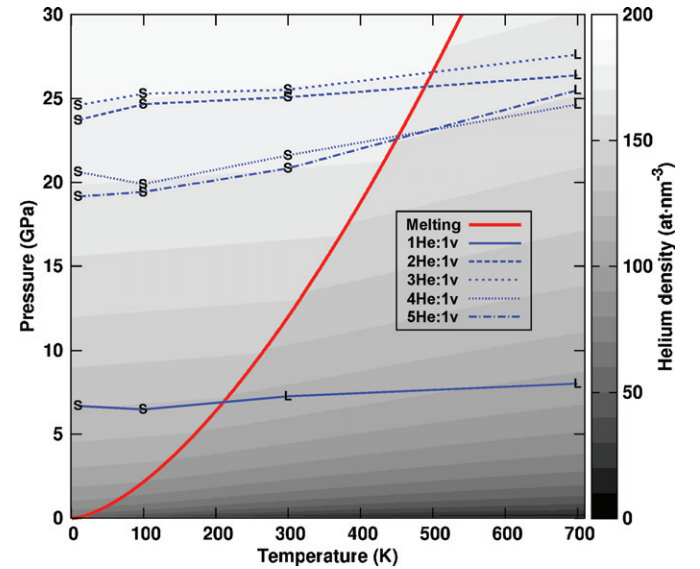


Fig. 4: (Colour on-line) Helium phase diagram and phase state of He in a 2 nm bubble in Fe as a function of He content in the temperature range from 10 to 700 K. The solid (left) to liquid (right) transition curve and He density contours are given according to Mills [29] and Trinkaus [22], respectively. “S” (solid) and “L” (liquid) symbols are the MD results.

are given in fig. 3, are obtained from the slope of the MSD *vs.* time graph. A self-diffusion coefficient of about 5×10^{-6} cm²·s⁻¹ in the temperature range from 10 to about 300 K is observed for all He contents. This corresponds to the self-diffusion coefficient of He in solid state [28]. When the temperature is increased beyond about 300 K the self-diffusion coefficient suddenly increases, which indicates that He liquefies.

From these observations a phase diagram of He in the bubble as a function of He content and temperature is derived (fig. 4). In this diagram “S” and “L” symbols are the MD results and refer to the solid and liquid state

of He, respectively. The analytical solid-liquid transition diagram, reported in fig. 4, obtained by Mills [29] gives the equation, which has been fitted to the Benedict-type equation of state according to the experimental observations. It appears that MD simulation results agree qualitatively well with it. The density calculated using the equations of state for the solid and liquid He derived by Trinkaus [22] is also reported in fig. 4. There is a good agreement with the atomic density obtained from MD calculation, fig. 2, at the specific He content and temperature. It appears thus that helium confined in a bubble in bcc-Fe behaves similarly to pure pressurized helium, despite the nanometric size and crystalline surrounding.

Beck potential was used to obtain the cohesion energy at 0 K for the fcc, hcp and bcc phases of He as a function of atomic density. Using this potential, the bcc structure appears to be the most stable structure, at an equilibrium atomic volume of around $18.5 \text{ \AA}^3$ per atom, although the energy difference between the bcc structure and fcc or hcp structures is small, at less than 10^{-4} eV per atom. When considering the helium density in the investigated He bubbles, however the fcc and the hcp structures are more favorable than the bcc structure, by about 10^{-3} eV per atom. According to a review [5] the fcc structure should be the stable one in the same pressure and temperature range.

The structure of He in the bubble is examined using the Voronoi common-neighbor analysis (CNA) [30]. It appears that, contrary to the above-mentioned observations, the structure of He is amorphous for all considered temperatures and pressures. However, some ordering is observed for the He atoms neighboring the iron atoms, as seen in fig. 5(a). They represent 10 to 20% of the bubble, and exhibit an icosahedral structure. The situation can be rationalized as follows. Van der Waals force in He is the weakest among all substances [31], which leads to easy disruption of its crystalline order. In addition, the lattice mismatch with surrounding bcc-Fe hinders the nucleation of He crystal. Also, the observed confinement of He atoms by the surrounding Fe lattice reduces the freedom of He atoms to crystallize.

The presence of the Fe crystalline structure around the bubble not only influences the structure of He but also impacts the bubble shape. MD simulation shows that at low He contents the bubble forms a rhombic dodecahedron with $\{110\}$ planes and $\{100\}$ truncations after annealing at 10 K, as depicted in figs. 5(b)–(d) for low He content bubble. Previous studies have shown that the critical parameter for the shape of a void is its surface energy. Nanometric void tend to polyhedral shapes having facets of low surface energy [32]. This was experimentally proven in bcc-Fe, where it forms a polyhedron with $\{110\}$ facets and $\{100\}$ truncations [33], as $\{110\}$ and $\{100\}$ surfaces have a lower energy of $2.3 \text{ J} \cdot \text{m}^{-2}$ than $\{111\}$ ones with $2.5 \text{ J} \cdot \text{m}^{-2}$ [23,24]. However, observation along different directions indicates that the 4He:1v bubble, fig. 5(e), tends to a perfect

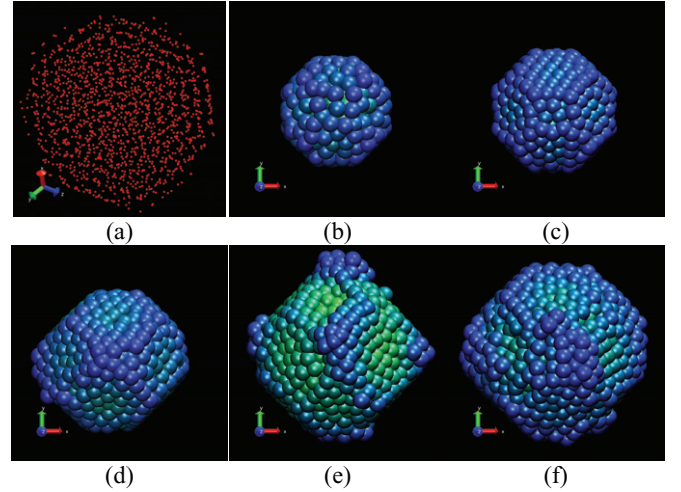


Fig. 5: (Colour on-line) a) He atomic configuration inside the 5He:1v 2 nm bubble in Fe lattice, and the morphology of the b) 1He:1v, c) 2He:1v, d) 3He:1v, e) 4He:1v and f) 5He:1v bubbles at 10 K. The axes correspond to $\langle 100 \rangle$ directions. In (a) the (red) points represents He atoms. In (b)–(f) green (light) to blue (dark) spectrum represents the radial distance of each atom from the bubble centre. Fe atoms are not shown here for clear depiction of the He bubble.

rhombic dodecahedron formed by $\{110\}$ planes and $\langle 111 \rangle$ edges. The corners extend in $\langle 100 \rangle$ and $\langle 111 \rangle$ directions. The largest extension of the bubble is in the $\langle 100 \rangle$ directions and the smallest extension is in the $\langle 111 \rangle$ directions, which are known to be the softest and hardest directions in bcc metals, respectively [34,35]. Moreover, beyond the 3He:1v ratio, when interstitial generation and plastic deformation starts, sharply defined $\{110\}$ facets are created because of the shear deformation on $\{110\}$ planes, which is known to be soft for shearing in bcc metals [36]. Increasing the He content beyond 4He:1v further deforms the bubble and some $\{100\}$ truncation appear once more, as shown in fig. 5(f). This indicates that He at high content behaves as a hydrostatic medium, as the bubble shape is dictated by the elastic and plastic properties of Fe. This behavior may contradict the fact that He in the bubble is in a solid state. Actually, according to *ab initio* calculations, solid He is a quasihydrostatic pressure-transmitting medium up to 150 GPa [37], which can thus just convey the bubble pressure to the surrounding Fe lattice. This explains the formation of the 12-faced rhombic dodecahedron in the case of the 4He:1v bubble.

Molecular-dynamics simulations reveal that a perfect iron crystal can hold with enough strength the He atoms in solid or liquid state within a nanometric bubble up to the theoretical strength of the perfect Fe crystal. It is expected that this behavior be found in other materials upon introduction of different gases provided that the gas solubility in the material is low enough to fulfill the conditions of bubble pressurization. We show that although the surface

energy criterion explains the formation of a polyhedron shape at low He content bubbles, the elastic and plastic properties of the surrounding material are dominant parameters in defining the shape of the bubble at high He contents, when pressure is high.

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