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# Self-trapping of helium in metals

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Atomistic calculations are presented which demonstrate that helium atoms in a metal lattice are able to cluster with each other, producing vacancies and nearby self-interstitial defects. Even a small number of helium atoms is found to be sufficient to create these large distortions. As few as five interstitial helium atoms can spontaneously produce a lattice vacancy and nearby self-interstitial. An eight-helium-atom cluster gives rise to two such defects, and 16 helium atoms to more than five self-interstitial vacancy pairs. It was noted that the self-interstitials prefer to agglomerate on the same "side" of the helium cluster rather than to spread themselves out uniformly. The binding energy of each additional helium atom to these clusters increases with helium concentration and the trap is apparently unsaturable. A rate theory using these atomistic binding energies has been used to calculate the kinetics of helium-bubble nucleation and growth. The results are consistent with measurements of the properties of helium resulting from tritium decay.

## I. INTRODUCTION

Helium atoms have long been known to become deeply trapped in metals when implanted at energies sufficient to create vacancies which the helium atoms can occupy. The helium, if implanted at below damage threshold energies, is mobile (depending on the temperature) as an interstitial and is therefore free to probe traps or defects which may have been deliberately introduced via a preimplantation phase of the experiment. Kornelsen used this effect in an extensive series of early experiments aimed at understanding helium trapping in metals following other rare-gas implantations.[1] Quantitative agreement was obtained between these measurements in tungsten[2] and atomistic calculations.[3]

The group at Delft (Caspers, van Veen, and coworkers) has also been actively pursuing helium-binding effects in implanted metals. Their experiments on Mo involved a preimplantation phase of 1-keV (above damage threshold) helium followed by low-energy (subthreshold) helium implantation and thermal desorption.[4] The appearance of three new peaks in the desorption spectra as the (subthreshold) helium dose was increased led these workers to postulate a "trap-mutation" model. In this model, existing vacancies containing helium ($\mathrm{He}_N V$) could mutate into divacancies ($\mathrm{He}_N V_2$) by emitting an isolated self-interstitial ($\mathrm{He} + \mathrm{He}_6 V \rightarrow \mathrm{He}_7 V_2 + I$). Their atomistic calculations[4] were unable to corroborate this trap-mutation model using the interatomic potentials of Wilson and Johnson.[5]

Recently, Kornelsen and van Gorkum[6] studied trapping of low-energy helium in tungsten preimplanted with He, Ne, Ar, Kr, and Xe at energies above threshold. They found that the helium binding to the rare-gas defects decreased monotonically with increasing rare-gas mass. Furthermore, these defects were found to continue to trap additional helium atoms at levels of more than 100 helium atoms per defect. Evans, van Veen, and Caspers[7] showed experimentally that helium platelets form after implantation of (damaging) 3-keV helium ions followed by (nondamaging) 150-eV implants.

When damage is not introduced, however, as is the case with low-energy helium implantation or with $^3$He introduction through tritium decay (sometimes called the "tritium trick" where the more soluble tritium gas enters the metal at elevated temperatures and is quenched to liquid-nitrogen temperature and aged, $\mathrm{T} \rightarrow {}^3\mathrm{He} + \beta^- + \nu$), one expects that the mobile helium interstitial will simply diffuse out of the sample (in the absence of impurities) at an appropriate temperature. Indeed, Kornelsen reports that at near-liquid-nitrogen temperatures in W, low-energy helium ions are not retained in the solid.[8] Furthermore, Wagner and Seidman[9] report that subthreshold He in W is mobile at ~96 K. The tritium measurements of Thomas, Swansiger, and Baskes,[10] however, indicate that ~98 at. % of the $^3$He generated in metal lattices remains in the lattice upon heating to temperatures as high as 500 °C. This retention occurs for cold-worked and polycrystalline Ni samples as well as for single crystals. Furthermore, the small release at low temperatures during their desorption experiments was found to be greater for those samples with lower initial concentrations (less aging) of $^3$He. These experiments point to a mechanism of helium retention independent of radiation damage.

These fundamental helium studies are made all the more relevant by the recent discovery of low-temperature helium embrittlement by West and Rawl.[11] These workers find a significant reduction in the ductility of steel containing both helium and tritium over those containing hydrogen alone. In fact, they report that the helium is from three to four times more effective, atom for atom, than hydrogen in degrading structural materials.

Here we present a self-trapping model of helium clustering and bubble growth in metals which ties together these experimental findings. Our intention is to build a model of helium behavior consistent with detailed atomistic considerations and with experiments. A rate-theory model, similar to one previously published,[12,13] has been used to compare the self-trapping results to helium behavior obtained from tritium-trick measurements. First indications of self-trapping were reported upon at the Dayton tritium conference.[14]

In Sec. II our atomistic and rate-theory calculations are described and results given. Section III contains a summary and discussion.

## II. ATOMISTICS OF SELF-TRAPPING

The method of calculation has been described in detail previously.[12,15] Briefly, a movable region of $N$ metal atoms and $M$ helium atoms is surrounded by a fixed zone of metal atoms and the potential energy (calculated as a sum of pair potentials) of the system as a function of the positions of these movable atoms is minimized using the conjugate gradients procedure. For the helium-helium interaction we use the potential derived by Beck[16] which was fitted to both *ab initio* theoretical as well as experimental data over a wide range of interatomic separations. The Ni-He potential has been derived by *ab initio* cluster calculations[17] and, as such, gives us a reasonable degree of confidence also.

The number of movable atoms was equal to 2093 for most of the calculations; allowing 8583 atoms to relax had a negligible effect on the results.[18] Two basic types of Ni-Ni potential were employed: (a) a long-range interaction due to Baskes and Melius[19] consisting of splines fitted to experimental data such as sublimation energy and vacancy formation energy as well as the stacking fault energy, and (b) a short-range form due to Johnson for Ni.[20] The long-range interaction has been fitted to a wider range of experimental data than the short-range potential (which originally was derived for $\alpha$-Fe). As will be discussed shortly, convergence problems were serious in these calculations, and the long-range form behaved better from this pont of view. We therefore report here the results for the long-range potential (making this set identical to potential set I of Ref. 14) although a large number of the calculations were also performed using the Johnson Ni-Ni potential. The essential features of the calculations are unchanged by this variation of potential. It should also be noted that in Fig. 7 of Ref. 14 we demonstrated that for five different choices of potentials, the Frenkel-pair-formation energy was significantly reduced as the number of helium atoms in a cluster increased.

These calculations were made difficult by the necessity of searching the very complicated $3(N+M)$-dimensional potential-energy surface. We found a large number of metastable configurations not reported here, configurations to which the energy converged very rapidly and smoothly. This was due to the large motions of the helium and metal atoms from their initial sites. The $\mathrm{He}_4$ (a cluster of four helium atoms) configuration reported at the Dayton conference[14] is a case of point: A lower-energy configuration having different symmetry has since been found and is reported here. Convergence problems were found to be more difficult for the short-range Johnson potential.[20] The results reported here therefore represent a great many calculations using many starting coordinates and we can offer no guarantee of their being absolute minima.

In Fig. 1 (and also in the Appendix), the energy required to bind the $N$th helium atom in a cluster of $N$ He atoms ($E_B$), that is, the binding energy required for the reaction $\mathrm{He}_N \rightarrow \mathrm{He}_{N-1} + \mathrm{He}$, is given for up to 20 He atoms. The detrapping energy is given approximately by the sum of this binding energy and the migration energy for interstitial motion ($E_D = E_B + E_a$). These energies were derived from end-point considerations alone, individual saddle points were not determined. The results are reported for clusters which initially contain no vacancies ($\mathrm{He}_N$). [See the Appendix for results involving one vacancy ($\mathrm{He}_N V$) and two va-

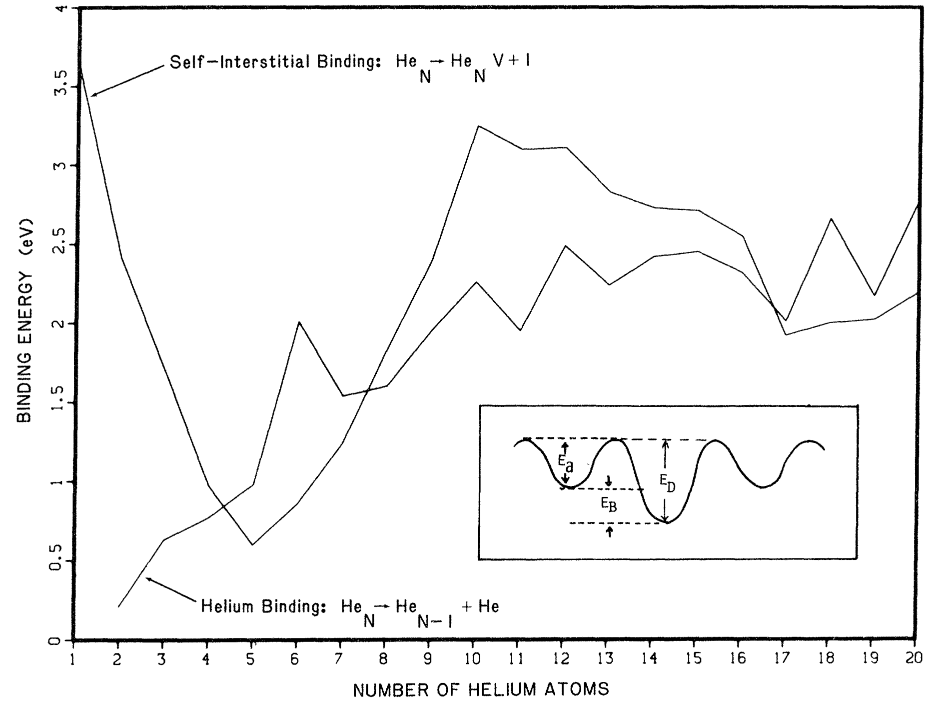

FIG. 1. Helium and self-interstitial binding energies as a function of the number of helium atoms in the cluster. The curves pertain to the case of no initial vacancies. Straight lines connect the discrete points. The detrapping energy ($E_D$) is given as the sum of the binding ($E_B$) and migration ($E_a$) energies as defined in the inset sketch.

cancies ($He_N V_2$).] As we shall see, the collective action of even small clusters of helium atoms is sufficient to spontaneously create additional vacancies and associated self-interstitials. That is, the helium can push lattice atoms off their normal sites. These lattice atoms, however, energetically prefer to remain in the near vicinity of the cluster resulting in a "near-Frenkel-pair" defect.[14]

The curve labeled "helium binding" in Fig. 1, then, gives the energy required to bind the $N$th helium atom in a cluster of $N$ helium atoms. Note that for the smallest cluster ($N = 2$) the binding is weak ($\sim 0.2$ eV) but that, as more helium atoms are added to the cluster, this binding increases to a rather high and near constant value above 2 eV. Thus, if such clusters were able to form in the lattice, the helium would be deeply trapped until the sample was exposed to very high temperatures. What is not obvious from this curve is the tremendous lattice "damage" or distortions introduced by helium. In Fig. 2 we have sketched the helium atoms and nearest surrounding Ni atoms for the $He_5$, $He_8$, $He_{11}$, and $He_{16}$ clusters (no initial vacancies). The (computer-drawn) sketches show the relaxed Ni atoms connected to their original lattice sites as a "bond" drawn to that site. The $He_5$ cluster has been shown earlier.[14] The remarkable feature of the distortions is that they are introduced by so few helium atoms. The $He_8$ cluster gives rise to two near-Frenkel pairs and the $He_{16}$

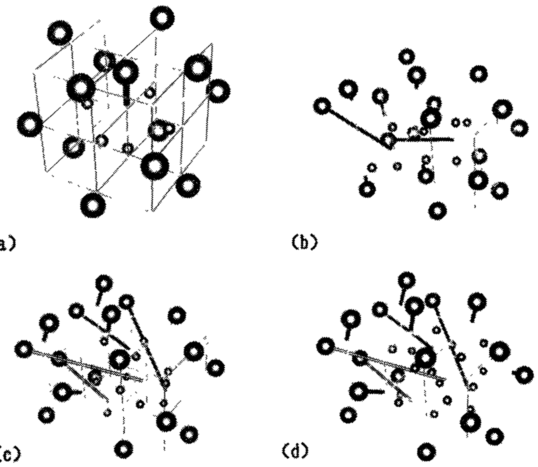

FIG. 2. Computer-drawn sketches of the minimum-energy configurations of (a) $He_5 \rightarrow He_5 V^* I^*$; (b) $He_8 \rightarrow He_8 V_2^* I_2^*$; (c) $He_{11} \rightarrow He_{11} V_7^* I_7^*$; and (d) $He_{16} \rightarrow He_{16} V_{10}^* I_{10}^*$ (see the Appendix) without introducing initial vacancies into the calculation. Near-Frenkel pairs are denoted $V^* I^*$.

cluster to more than five of these defects, depending upon one's definition of how far a lattice atom must be displaced from its normal site to be considered an interstitial. An extended discussion and further calculations concerning this point are to be found in the Appendix.

One feature of the minimum-energy geometry of these clusters, which has physical significance, is that the self-interstitials always prefer to cluster on the same "side" of the embryonic helium bubble. This preference led us to an important digression: the calculation of the self-interstitial clustering energies in the *absence* of helium. We find a very strong propensity for these defects to agglomerate due to the resultant reduction in elastic strain in agreement with the results of Ingle, Perrin, and Schober.[21] The detailed energies and geometries of these clusters will be presented in a later paper; the importance of this clustering phenomena to this work is that self-interstitials produced by the helium clusters energetically prefer to cluster on one side of the defect (Fig. 2) rather than to distribute themselves more or less uniformly on the "surface" of the defect. It is this clustering of self-interstitials in the near region of the helium cluster which makes self-trapping energetically favorable in a number of cases. Because the Delft group[4] omitted this stabilizing effect, their calculations were found to disagree with their experimental suggestion of mutation. This experimental suggestion may now be seen to be correct. Most importantly, self-interstitial stabilization provides a mechanism for bubble growth without requiring the self-interstitials to detrap.

The detrapping of these bound self-interstitials is a competing process to the detrapping of helium atoms. In Fig. 1, the binding energy for this process is given as a function of the number of helium atoms in the cluster when no initial vacancies are present. The helium initially (small number of atoms) aids the detrapping of the self-interstitial (i.e., mutation) by introducing a repulsive force to the metal atom as discussed above. For large clusters, however (specifically beyond $He_5$), the self-interstitial finds it even more energetically preferable to remain in the strain field of the cluster rather than to form an isolated interstitial. This introduces the strong minimum in the self-interstitial binding curve. The additional structure in the figure is caused by geometrical factors and also by the punching out of additional near-Frenkel pairs (see the Appendix).

From these calculations, a qualitative picture of the physical mechanism of cluster growth without radiation damage can be constructed. At low concentrations of $He << 1$ ppm (short aging times in a tritium experiment) or short diffusion paths, mobile helium interstitials are unlikely to experience each other's strain field in their diffusion out of the sample and hence clustering does not take place. For example, a He atom samples $\sim 10^7$ sites in a random walk of 100 $\mu$m; hence it encounters another He atom during this random walk only for He concentrations $\gtrsim 0.1$ ppm. As the concentration of helium is increased, the weak ($\sim$0.2 eV) binding of one He atom to another He (formation of $He_2$) can delay the helium interstitial until a third helium migrates along and is trapped more deeply ($\sim$0.6 eV). A fourth helium may then join the third and a fifth join the four-atom cluster at which point a lattice "vacancy" (near-Frenkel pair) is spontaneously produced. In addition to temperature (helium mobility), the concentration necessary for this trapping to occur also depends upon the distance to other sinks (e.g., surfaces or impurities). At a critical number of clustered interstitial helium atoms ($He_5 \rightarrow He_5 V^* I^*$), a near-Frenkel pair (denoted $V^* I^*$) is produced and the sixth helium is then more deeply bound than the fifth. However, at high enough temperature, the competing process of self-interstitial detrapping may take place from Table II [0.61 eV + 0.15 eV ($E_{1I}^{M}$)=0.76 eV] transforming the cluster into an $He_5 V$. This process occurs near the crossing of the helium binding and interstitial binding curves of Fig. 1 but must be corrected by adding the helium and self-interstitial activation energies for migration to the binding energies. The next self-interstitial ($He_5 V \rightarrow He_5 V_2 + I$) is then deeply bound (2.78 eV, Table II) until the cluster gains five more helium atoms ($He_{10} V$) at which point the self-interstitial ($He_{10} V \rightarrow He_{10} V_2 + I$) is essentially free to migrate (0.32 eV binding) into the lattice.

As a specific example of these clustering phenomena we have explicitly modeled the tritium decay measurements of Thomas *et al.*[10] The actual physical processes that take place in this experiment are complicated by such parameters as the solubility of tritium in the lattice. During a tritium-trick experiment, the sample may experience a room-temperature anneal for a long enough time period for tritium to diffuse out of the near-surface regions of the sample. The $^3$He concentration, born from this tritium, would then be a function of depth in the sample and hence the character of the helium clusters which form during thermal desorption would also be a strong function of depth.

In order to handle such complications directly and in order to demonstrate that the helium self-trapping described here actually occurs in real physical systems, a coupled set of rate equations was solved which utilized the binding energies presented in Tables I and II. The mathematical technique has been published previously[12,13] and further details will be described in a forthcoming paper. Briefly, a system of stiff, coupled first-order partial differential equations allowing both diffusion and the trapping and detrapping of helium, self-interstitials, and vacancies was solved numerically. The experimental conditions corresponding to the (Ni) tritium-trick experiments of Thomas *et al.*[10] were employed including the calculated tritium distribution near the surface. The calculations included $\mathrm{He}_N V_M$ clusters for $N=1$ to 20 and $M=0$ to 2 and were performed for a variety of helium-diffusion activation energies.

The potentials employed here give rise to a helium activation energy of $\sim 0.6$ eV as reported earlier.[19] A more flexible hybrid method gave 0.43 eV (Ref. 17) as the helium activation energy in Ni while analysis of the experiments[10] indicated a still lower value of 0.35 eV. As we have stated many times, defect calculations are valuable in giving trends and essential processes and should not be expected to give absolute quantitative agreements. The values of the binding energies in the tables could well be changed by $\sim 0.2$ eV or more by the use of still another potential or the inclusion of other effects (such as *d* electrons explicitly). Our variation of the potentials gives us a rough estimate of the accuracy of the calculation.

Taking the values of the binding energies given in the tables at face value, we calculated the amount of helium evolved from the sample during the same linear ramp anneal employed in the tritium-trick measurements (10 K/min). The fractional amount of helium released by room temperature is plotted as a function of helium interstitial activation energy in Fig. 3 for two initial helium concentrations. (The concentrations of clusters of all types formed as a function of depth, etc., will be presented in a future publication.) Note that, in agreement with experiment ($\lesssim 2\%$ helium released), very little helium is released if the helium interstitial activation energy $E_{\mathrm{He}}^M$ is below $\sim 0.3$ eV.

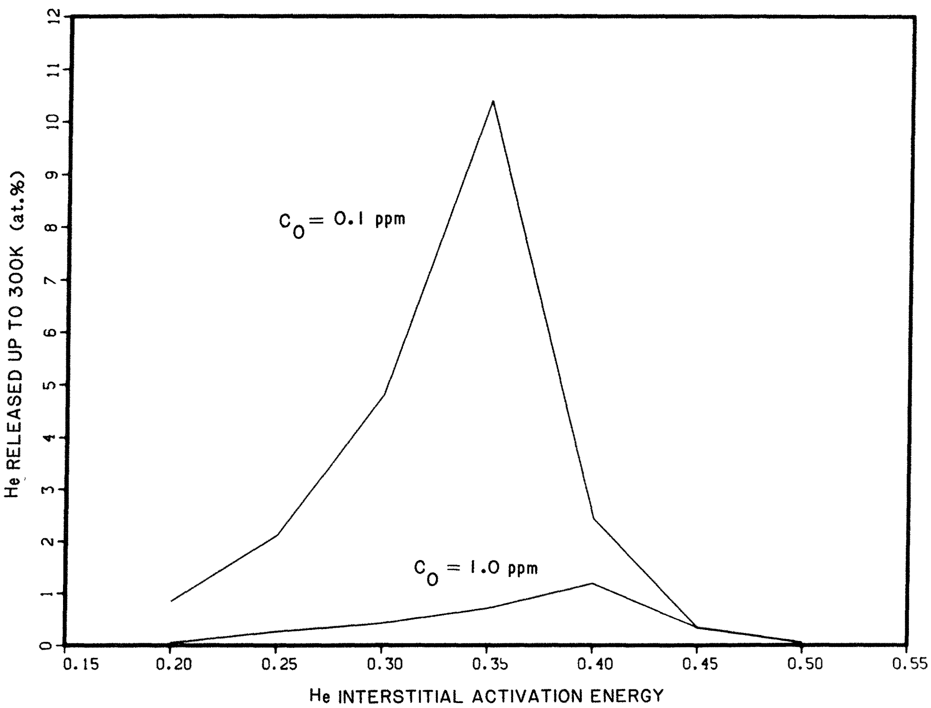

FIG. 3. The fractional release of $^3$He in a tritium-trick experiment using the binding energies from Tables I and II for initial $^3$He concentrations, $C_0$, as a function of helium interstitial activation energy. Note the reduced fraction of helium release in the sample containing the larger initial concentration, in agreement with experiment.

At small interstitial helium activation energies, the highly mobile helium is able to form large clusters during the early stages of the linear ramp, hence the fraction released during thermal ramping is small. At higher activation energies the cluster growth is delayed to higher temperature (due to lower helium mobility) and some helium is able to detrap (see Fig. 1) below room temperature. For example, our calculations show that a helium interstitial activation energy of 0.25 eV gives rise to an average of 6.1 He atoms per cluster while an activation energy of 0.30 eV yields an average of 1.2 He atoms per cluster. From Fig. 3 one notes the rapid increase of helium released in this activation energy range. The reader should note from Table I that the second helium is bound by only 0.2 eV while the sixth is bound by $\sim 2$ eV.[22] At higher helium interstitial activation energies, the helium is unable to diffuse out of the lattice at temperatures at or below room temperature. Hence, a peak results in the curves of Fig. 3. Note also that the concentration effect (relatively more helium is released when less is available initially) also reproduces the trend of the experiment. Furthermore, as will be shown in a forthcoming paper, an activation energy of 0.3 eV gives rise to a peak in the ($<2\%$) low-temperature release rate at $\sim 190$ K, again in agreement with experiment.

## III. SUMMARY AND DISCUSSION

We have shown using atomistic calculations that small clusters of helium atoms produce enormous lattice distortions and lead to the formation of near-Frenkel pairs (lattice atoms pushed into nearby interstitial sites). We find the critical number of helium atoms necessary for these processes to occur is very small ($\mathrm{He}_5 \rightarrow \mathrm{He}_5 V^* I^*$; $\mathrm{He}_8 \rightarrow \mathrm{He}_8 V_2^* I_2^*$). One important feature of the cluster geometry is the energy lowering we find when the self-interstitials, pushed from their lattice sites, themselves cluster on one side of the helium bubble to reduce the strain energy. We find, in agreement with the recent work of Ingle *et al.*,[21] that self-interstitials in Ni favor clustering in the absence of helium.

The calculated binding energies were employed in a model, based on rate theory (further details to be published later), which was found to agree semiquantitatively with the tritium-trick measurements of Thomas, Swansiger, and Baskes.[10] In addition, the model is consistent with the low-energy implants of Thomas and Bastasz[23] as well as the embrittlement found by West and Rawl.[12] Helium clusters produce self-interstitials which form loops and strengthen the material in much the same way as solid solution strengtheners. The grain boundaries then yield before the bulk material.

Clearly, if homogeneous nucleation as shown here can occur, the introduction of traps such as impurities or inclusions in stainless steel (or rare-gas atoms introduced deliberately to trap helium) can only enhance the trapping by acting as an additional initial nucleus for the helium cluster. The Kornelsen and van Gorkum[6] result is a case in point: Preimplantation of He, Ne, Ar, Kr, and Xe produced trapping sites for helium, the character of which became less identifiable as the concentration of (subthreshold) helium was increased. The results presented here indicate that helium trapping with the concurrent formation of self-interstitials is occurring in these experiments with the preimplant rare gas acting as a strong initial trap site. For example, He is more deeply trapped to an Ar substitutional than to a He interstitial. It is also to be noted that we find no saturation of these traps for the largest numbers of helium we have so far included (20), consistent with the results of Kornelsen and van Gorkum.[6]

In bcc materials, the helium interstitial activation energy is lower than in fcc materials.[5,9] Since the binding energies of helium interstitials to each other in bcc's are even greater than in fcc's,[12,24] it is clear that self-trapping will also occur in these materials given appropriate experimental conditions. Subthreshold implantation of W at slightly above liquid-nitrogen temperature results in the helium coming out as observed by Kornelsen[8] because the helium concentration never builds up to a high enough level to produce clustering. The only difference between fcc and bcc materials for these arguments is the increased interstitial mobility and deeper trapping for bcc's. We would predict self-trapping of subthreshold helium in bcc materials at implant temperatures low enough to prevent helium migration.

Finally, Baskes *et al.*[25] have shown using a Monte Carlo model that if helium can create lattice vacancies and if the resulting self-interstitials remain attached to the cluster, helium bubbles readily grow. Their assumption of one near-Frenkel pair created per helium atom (beyond a critical number of helium atoms) is not unreasonable.

## ACKNOWLEDGMENTS

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## APPENDIX

In Table I we present the results of our calculations of the binding energy of the *N*th helium atom in a cluster of *N* helium atoms (i.e., for the reaction $\mathrm{He}_N V_M \rightarrow \mathrm{He}_{N-1} V_M + \mathrm{He}$) for the long-range potential set described in the text. The calculations were performed including no initial vacancies ($\mathrm{He}_N$), one initial vacancy ($\mathrm{He}_N V$), and two initial vacancies ($\mathrm{He}_N V_2$). The first column of Table I is plotted in Fig. 1 of the text.

In Fig. 2 of the text, we have sketched the minimum-energy configurations for a few of the critical clusters. Here we attempt a numerical description of the number of near-Frenkel pairs (vacancy-interstitial pairs with the interstitial remaining bound in the vicinity of the vacancy) introduced by a given number of helium atoms. Just how far a lattice atom needs to be forced off its normal site to be considered an interstitial is, of course, arbitrary.

Using the lattice positions calculated in obtaining Table I above, we determined the number of lattice atoms at or beyond one half-lattice-constant (hlc) from their normal sites and also those at or beyond $\sqrt{2}/2$ hlc (i.e., one-half the first-nearest-neighbor distance). The results are presented in Table I. The difference between the two definitions becomes as much a factor of 2 (with respect to the number of Frenkel pairs created) as the number of helium atoms increases.

The $\mathrm{He}_5$ cluster, which we previously defined as creating a near-Frenkel pair[14] pushes a lattice atom along a ⟨100⟩ direction 0.72 hlc and hence this atom is not defined as an interstitial in our first definition despite its enormous motion. A small local minimum occurs at this position. From Table II

TABLE I. Binding energies of helium to helium clusters containing no initial vacancies and also one and two initial vacancies. The minimum-energy configurations of the clusters are also given by defining a near-Frenkel pair as occurring when the self-interstitial has been displaced at least one-half lattice constant (hlc) from its normal site. Configurations given in parentheses use a $\sqrt{2}/2$ hlc definition for existence of a near-Frenkel pair.

| Number of helium atom, *N* | No vacancies $\mathrm{He}_N \rightarrow \mathrm{He}_{N-1} + \mathrm{He}$ | | One vacancy $\mathrm{He}_N V \rightarrow \mathrm{He}_{N-1} V + \mathrm{He}$ | | Two vacancies $\mathrm{He}_N V_2 \rightarrow \mathrm{He}_{N-1} V_2 + \mathrm{He}$ | |
|---|---|---|---|---|---|---|
| | Binding energy (eV) | Number of near-Frenkel pairs | Binding energy (eV) | Number of near-Frenkel pairs | Binding energy (eV) | Number of near-Frenkel pairs |
| 1 | | | 2.63 | 0(0) | 2.83 | 0(0) |
| 2 | 0.22 | 0(0) | 1.44 | 0(0) | 2.85 | 0(0) |
| 3 | 0.64 | 0(0) | 1.35 | 0(0) | 1.98 | 0(0) |
| 4 | 0.78 | 0(0) | 1.51 | 0(0) | 1.84 | 0(0) |
| 5 | 0.99 | 0(1) | 1.35 | 0(0) | 1.84 | 0(0) |
| 6 | 2.02 | 1(1) | 1.76 | 0(0) | 1.40 | 0(0) |
| 7 | 1.55 | 1(1) | 1.16 | 0(0) | 2.05 | 0(0) |
| 8 | 1.60 | 2(2) | 1.02 | 0(0) | 1.63 | 0(0) |
| 9 | 1.95 | 2(2) | 1.38 | 0(0) | 2.09 | 0(0) |
| 10 | 2.27 | 6(9) | 1.41 | 0(0) | 2.04 | 0(0) |
| 11 | 1.96 | 5(7) | 2.11 | 5(7) | 1.35 | 0(0) |
| 12 | 2.50 | 5(7) | 2.49 | 5(7) | 1.44 | 0(0) |
| 13 | 2.25 | 5(7) | 2.53 | 5(7) | 2.29 | 0(0) |
| 14 | 2.42 | 5(7) | 2.53 | 5(7) | 1.63 | 5(7) |
| 15 | 2.45 | 5(9) | 2.47 | 5(9) | 2.57 | 5(7) |
| 16 | 2.33 | 5(10) | 2.48 | 6(10) | 2.51 | 5(7) |
| 17 | 2.02 | 5(11) | 2.65 | 6(10) | 2.81 | 5(7) |
| 18 | 2.66 | 6(11) | 2.58 | 6(10) | 2.51 | 5(8) |
| 19 | 2.18 | 5(10) | 2.16 | 9(11) | 2.33 | 5(9) |
| 20 | 2.78 | 6(12) | 2.62 | 6(13) | 2.70 | 5(10) |

we see that only 0.61 eV ( + 0.15 eV for self-interstitial migration) is required to remove this atom to an isolated position in the lattice.

For $N \geq 11$, we find several lattice atoms are displaced far from their lattice-sites (see Table I) regardless of the definition of self-interstitial formation. Since a self-interstitial can migrate in the lattice for only $\sim 0.15$ eV, a very small energy barrier needs to be overcome. The same low-energy barrier seems to apply to collective motions of these interstitials, making it difficult to distinguish between configurations or to unambiguously define a minimum energy cluster. For example, the formation energy of $\mathrm{He}_{10}V_2^*I_2^*$ is only 0.1 eV higher than $\mathrm{He}_{10}V_4^*I_4^*$ and $\mathrm{He}_{10}V_6^*I_6^*$ is 0.2 eV above $\mathrm{He}_{10}V_4^*I_4^*$. (Vacancies and interstitials which remain in the vicinity of the cluster are identified with an asterisk.) That is, the motion of several lattice atoms off their sites due to the enormous ten-helium-atom strain field has only a very small effect on the energy. The reader is therefore cautioned not to apply too rigorous a definition to the minimum-energy configurations we found.

Returning to Table I, we note that a minimum exists in the curves of the binding energy versus the number of helium atoms for clusters containing an initial isolated vacancy $\mathrm{He}_N V$, or divacancy $\mathrm{He}_N V_2$. For $\mathrm{He}V$, $\mathrm{He}V_2$, and $\mathrm{He}_2V_2$ the vacancy is essentially acting as a "hole" for the helium. (The interstitial helium formation energy is 4.02 eV and it requires only 1.39 eV to place the helium atom in an existing vacancy.) As the number of helium atoms increases, the repulsive He-He force lowers the binding energy of the helium (increases the formation energy for helium in the helium-containing vacancy). For $\mathrm{He}_6V$ the peak noted earlier[12] appears due to the high symmetry of this defect. This peak is shifted to seven helium atoms when two vacancies are involved ($\mathrm{He}_7V_2$) as might be expected. The binding energy continues to decrease until at $\mathrm{He}_8V$ ($\mathrm{He}_{11}V_2$) a minimum is reached as a result of the "punching" out of additional vacancies. Hence, $\mathrm{He}_8V$ actually becomes $\mathrm{He}_8VV^*I^*$ which further binds the helium. Similarly, $\mathrm{He}_{11}V_2$ spontaneously forms $\mathrm{He}_{11}V_2V^*I^*$. At the largest numbers of helium atoms considered here, the binding energy again levels out at an asymptotic value (e.g., an average of the last five highest energies) which seems to increase slightly with the number of vacancies present.

In Table II we report the binding energies of the self-interstitials to the helium clusters for the cases

TABLE II. Binding energies of self-interstitials to helium clusters containing no initial vacancies and one initial vacancy.

| Number of helium atoms, $N$ | No vacancies $\mathrm{He}_N \rightarrow \mathrm{He}_N V + I$ | One vacancy $\mathrm{He}_N V \rightarrow \mathrm{He}_N V_2 + I$ |
|---|---|---|
| 0 | 6.27 | 5.83 |
| 1 | 3.64 | 5.64 |
| 2 | 2.42 | 4.22 |
| 3 | 1.71 | 3.60 |
| 4 | 0.97 | 3.27 |
| 5 | 0.61 | 2.78 |
| 6 | 0.86 | 3.14 |
| 7 | 1.25 | 2.25 |
| 8 | 1.83 | 1.65 |
| 9 | 2.40 | 0.94 |
| 10 | 3.26 | 0.32 |
| 11 | 3.11 | 1.07 |
| 12 | 3.12 | 2.12 |
| 13 | 2.84 | 2.35 |
| 14 | 2.73 | 3.25 |
| 15 | 2.71 | 3.16 |
| 16 | 2.56 | 3.12 |
| 17 | 1.93 | 2.96 |
| 18 | 2.01 | 3.03 |
| 19 | 2.03 | 2.86 |
| 20 | 2.19 | 2.78 |

of no initial vacancies and one initial vacancy. The first column of the Table is plotted in Fig. 1. There is a minimum in the binding energy of the self-interstitial versus the number of helium atoms when an initial vacancy is included for the same reasons discussed in the text (in the absence of the vacancy). However, the minimum is shifted to 10 helium atoms and happens to have a lower value than for the first interstitial. Further calculations of the repetition of this process are underway.

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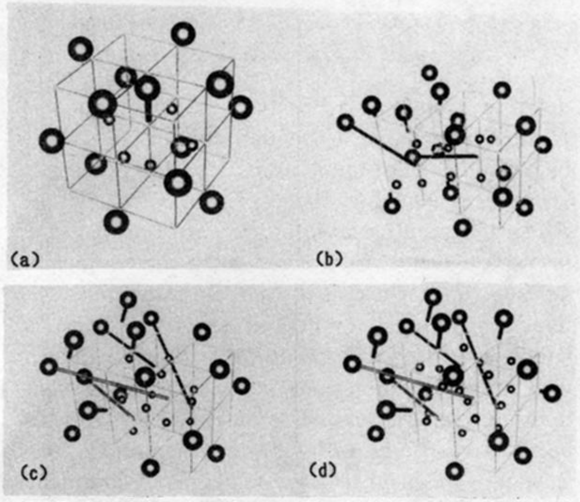

FIG. 2. Computer-drawn sketches of the minimum-energy configurations of (a) $\mathrm{He}_5 \rightarrow \mathrm{He}_5 V^* I^*$; (b) $\mathrm{He}_8 \rightarrow \mathrm{He}_8 V_2^* I_2^*$; (c) $\mathrm{He}_{11} \rightarrow \mathrm{He}_{11} V_7^* I_7^*$; and (d) $\mathrm{He}_{16} \rightarrow \mathrm{He}_{16} V_{10}^* I_{10}^*$ (see the Appendix) without introducing initial vacancies into the calculation. Near-Frenkel pairs are denoted $V^* I^*$.