In situ TEM observation of helium bubble evolution in V/Ag multilayer during annealing

Hongxiu Zhang ^a, Feng Ren ^{a,*}, Yongqiang Wang ^b, Mengqing Hong ^a, Xiangheng Xiao ^a, Wenjing Qin ^a, and Changzhong Jiang ^a

^a School of Physics and Technology, Center for Ion Beam Application and Center for Electron Microscopy, Wuhan University, Wuhan 430072, P. R. China

^b Materials Science and Technology Division, Los Alamos National Laboratory, Los Alamos, New Mexico 87545, USA

* Corresponding authors. Tel: +86-27-68752567; Fax: +86-27-68752569. E-mail: fren@whu.edu.cn

Abstract

Multilayers have been considered as promising candidates for nuclear materials. In this paper, an in situ study on the growth of helium bubbles during annealing was performed by transmission electron microscopy. Bubbles in the Ag layers grew rapidly by vacancy absorption mechanism and reached a size comparable to the layer thickness, whereas bubbles in the V layers evolved slowly due to the lower diffusion rate of vacancies and stopped growing at the size of 1.8 nm. The interfaces with more than one orientation relationship in the V/Ag multilayer contained pockets of excess volumes that can readily trap helium and exhibit high helium solubility. The interfaces prevent bubbles from growing into nearby layers.

1. Introduction

A problem of particular concern in some of nuclear fission and fusion energy systems is the effects of helium on the microstructures and mechanical properties of materials [1-4]. Due to the extremely low solubility of He in metals (approximately parts per billion level), upon introduced by (n,a) either reactions in the reactor or by direct injection through ion implantation, He atoms strongly tend to combine with vacancies to form He-vacancy clusters, which act as the nucleus for the growth of He bubbles [5]. Once nucleated, the bubbles grow through absorbing the successive injected He atoms and irradiation induced vacancies in order to maintain a mechanical equilibrium between their internal He pressure and the stress originated from the surface tension of the bubble, $2\gamma/r$, where γ is the He/metal interface energy and r is the bubble radius [1, 2, 6-9]. He bubbles cause deleterious effects such as swelling and embrittlement of the materials. Therefore, controlling the formation and evolution of defects and He bubbles in nuclear materials is an important task for the materials research community.

Numerous researches have demonstrated that nano-structural materials are radiation tolerant and have the ability to handle radiation-induced defects and bubbles. Bai *et al.* [10] showed that grain boundaries could act as efficient sinks for both vacancies and interstitials through the “loading-unloading” mechanism. Zinkle and Singh [11] proposed grain-size dependent nucleation of defects in neutron irradiated alloys. Singh [12] showed that the formation of the ‘defect depleted zone’ along the grain boundary can reduce the growth rate of growing voids as the distance between the voids and grain boundaries decreases. Our own studies recently revealed that the V and CrN films with nano-channel structures also achieve great radiation tolerance through annihilation of freely-migrating point defects and release of He at free surfaces [13, 14].

Multilayers based on two alternating metallic films with high interfacial areal fractions have been considered as promising candidates for nuclear materials. A considerable amount of research has shown that interfaces in multilayers can act as sinks to store radiation-induced point defects, enhance the diffusivity of point defects along interfaces, promote the recombination of point defects, and thus improve the radiation tolerance of the multilayers [6, 10, 15]. Such concepts have been well proved in a number of multilayer systems including Cu/Nb [9, 16], V/Ag [3, 17], Fe/W [1], CrN/AlTiN [18] etc., in which, reduced He bubble density and reduced hardening were observed through decreasing periodic thickness in multilayered structure. Recently, Han et al. [19, 20] fabricated Cu/Nb multilayers through the accumulative roll bonding method, making multilayered nanofilms amenable to large-scale industrial production.

Ion irradiation is considered to be an efficient method for the evaluation of radiation effects in different materials as it can induce large numbers of defects such as interstitials, vacancies and defect clusters. Noble-gas-ion-irradiation-induced external species, such as He, easily combine with vacancies

to form nano-size or even larger sizes of bubbles [1, 2, 6, 8, 9, 21]. With its advantages in direct observation, accuracy and reliability, transmission electron microscopy (TEM) has become one of the most extensively used methods for characterizing irradiation-induced defects and He bubbles in nuclear materials. In situ TEM ensures the real time observation of the microstructural evolution of a material during thermal annealing or in situ ion irradiation. The information obtained from such studies is important not only for the fundamental understanding of gas bubble formation and growth in material but also for designing new radiation tolerant materials. Through in situ Kr^+ ion irradiation within a TEM, Sun et al [22] demonstrated that high angle GBs in nanocrystalline Ni can effectively absorb irradiation-induced defects. Using in situ TEM observation, Ran et al [23] found that the growth mechanism of the bubbles in a zirconium alloy is through surface diffusion. K. Ono et al. [24] observed the Brownian motion of He bubbles through in situ irradiation on Al foils. To the best of our knowledge, there has been no in situ TEM studies on the thermodynamic behavior of He in irradiated multilayers. Since the interfaces in multilayers are known to be efficient sinks for vacancies, interstitials and other kinds of defects [3, 18, 25-27], understanding the evolution of He bubbles in multilayered films under thermal annealing could provide important insights to assist in designing radiation tolerant materials.

As an immiscible system, the interfaces of the V/Ag multilayer are expected to be stable against radiation induced inter-mixing [28]. Previous experimental results by Wei and Misra showed that neighboring Ag and V layers had a variety of orientation relations [28]. In addition, the interfaces of V/Ag multilayers are incoherent since their lattice parameter mismatch is rather large (e.g. 8.9% in the Kurdjumov-Sachs (K-S) orientation relationship) [28]. Atomistic modeling on Cu/Nb multilayers suggested that misfit dislocation intersections (MDIs) in Cu/Nb multilayers contain pockets of excess volume that readily trap He [29]. It was also suggested that, due to the existence of more than one type

of orientation relationships between V and Ag layers, the V/Ag interfaces contain high density of MDIs. Moreover, the misfit dislocation spacing in the interfaces are of the order of a few nanometers suggesting that the interfaces possess a large number of defect trapping sites. Therefore, V/Ag multilayers are expected to have good radiation tolerance [3, 27]. In this paper, we report our in situ study on the growth behavior of He bubbles in the irradiated V/Ag multilayer during annealing in a TEM. To the best of our knowledge, this is also the first in situ TEM study of thermal-annealing-induced He bubble evolution in multi-layered structures.

2. Methods

A V/Ag multilayer film was deposited on HF-etched Si (1 0 0) wafers at room temperature by alternate deposition of V and Ag in an ultra high vacuum magnetron sputtering system (ULVAC, ACS-4000-C4). The vacuum chamber was evacuated to a base pressure of less than 5×10^{-6} Pa prior to deposition, and the deposition was then performed under a working pressure of 5×10^{-1} Pa. The working gas was Ar and the distance between the targets and the substrate was kept at 130 mm. Prior to the deposition of the V/Ag multilayer a 10 nm thick V layer was deposited as a buffer layer on the substrate. The last layer was V to avoid surface oxidation. The deposition rates were controlled at approximately 4 nm min^{-1} for V and 20 nm min^{-1} for Ag. The planned thicknesses of Ag and V layers were 6 nm each; but the actual thicknesses were 6.5 for Ag and 4.5 nm for V, respectively. The total thickness of the multi-layer was approximately 350 nm. After deposition, the multi-layer film was irradiated at room temperature with 40 keV He^+ ions to a fluence of $5 \times 10^{16} \text{ ions/cm}^2$ under a flux of $1 \times 10^{13} \text{ ions/cm}^2/\text{s}$. The He concentration and irradiation damage as a function of the penetration depth were calculated by the computer code SRIM 2013, which is based on the Monte Carlo method [30, 31].

The typical value of 25 eV was used as the displacement energy for both Ag and V and the full cascade mode was used in SRIM calculations. For the given He beam energy and fluence, the SRIM predicted that the peak concentration of He was ~5 at.% at a depth of 170 nm, assuming a density of 5.96 g/cm³ and 10.47 g/cm³ for V and Ag, respectively.

The irradiated film specimen was prepared for TEM observation. Sample preparation consisted of standard film cross-sectioning techniques: sample mounting between two Si wafers using Gatan M-bond 610 epoxy, mechanical polishing to approximately 40 μm by abrasive paper using the Gatan precision disc grinder, dimpling to approximately 20 μm , gluing on to a molybdenum grid and final thinning to electron transparency using a Gatan Precision Ion Polishing System. Ion milling with energetic Ar ions may produce artifacts including preferential sputtering, specimen heating and surface damage on the electron-transparent region of the sample. To reduce the damage, a low incident angle (3 degrees) and low energy (2.5-3 keV) Ar ions were applied to the final stage of the ion thinning process. The TEM sample was annealed from room temperature to 540 °C on a Gatan 652 double tilt heating holder in the JEM-2010 (HT) (HT: high angle tilt) TEM operated at 200 kV. The duration of the heating process was 5250 s. Videos and still images were captured by a Dualvision CCD digital camera. The frame size was 640×480 pixels and the frame rate of the videos was 6 Hz. The images of the as-deposited samples were captured under both in-focus and under-focus conditions. The images of the samples after He ion implantation were captured in an under-focus condition. Since the bubble image size is very sensitive to the under-focus conditions, especially for the bubble diameter around 1 nm, to get the best image contrast we kept the under-focus as constant as possible to -1100 nm by adjusting the current of the microscope objective lens during the annealing process. In the absolute sense, it is possible that the measured size of the bubbles may be slightly larger than the real value. For a given

fixed under-focus value, we believe that the bubble image sizes measured under different annealing temperatures still provide a reliable trend on the temperature dependence of He bubble growth in these samples.

3. Results and discussion

3.1. Interface trapping

Figure 1(a) shows the under-focus cross-sectional TEM micrograph of the as-deposited V/Ag multilayer with a polycrystalline structure. The selected area electron diffraction (SAED) pattern indicates that the multilayer film consists mainly of Ag (111) and V (110) planes parallel with the surface

To exclude the possibility of interface mixing, axial dark-field (DF) images were taken by selecting the Ag (111) diffraction spot marked in the SAED image, and the results indicate chemically sharp interfaces between Ag and V sublayers. Figure 1(b) is the HRTEM analysis of the V/Ag multilayer suggesting that the Kurdjumov-Sachs (K-S) orientation relationship with $\{111\}\text{Ag} // \{110\}\text{V}$ and $\langle 110 \rangle \text{Ag} // \langle 111 \rangle \text{V}$ is dominant in the V/Ag interfaces [28]. Other minor orientations were also observed including the pilsch relationship with $\{100\}\text{Ag} // \{110\}\text{V}$ and $\langle 110 \rangle \text{Ag} // \langle 111 \rangle \text{V}$. It should be noted that there is an angle of 5° between the $(1\bar{1}\bar{1})$ of Ag and $(\bar{1}0\bar{1})$ of V, which results in the deviation of the V $(\bar{1}0\bar{1})$ with the straight line along the Ag $(1\bar{1}\bar{1})$. At the interface where the Ag planes join the V planes, dislocations or disconnections are expected to form in order to relieve the misfit strain. The rotation of the two lattices was driven by the minimization of the misfit strain at the interface [28, 32].

Figure 2(a) shows under-focus cross-sectional TEM micrograph of the He-irradiated V/Ag

multilayer. The defocus was set at -1100 nm, the optimum imaging condition for our experimental setup. Figure 2 (a) shows that the V/Ag layered morphology was not degraded upon the high fluence irradiation of energetic He^+ ions. Furthermore, no formation of intermixing or amorphous layer was observed, which confirms the previous studies on immiscible V/Ag [17] and Cu/Nb [33] multilayers synthesized by magnetron sputtering after He ion irradiation. Since the irradiated systems consist of immiscible components, phase separation is expected to compete with ion irradiation-induced mixing. Averback et al. [34] studied the mixing behaviour of Cu/Nb system by 750 keV Kr ion irradiation, and concluded that ion beam mixing occurs at cryogenic temperature during ion implantation, but is greatly reduced when the ion irradiation process is carried out at room temperature. In addition, irradiation with light ions like He was found to only result in equilibrium phases [33]. Superimposed on the Fig. 2(a) is a depth profile of the irradiation damage obtained from the SRIM calculations, which predict that the peak damage region of 5 dpa is located at a depth of 150 nm below the surface. Notably, most of the bubbles formed in the Ag layers. Bubbles marked by the red circles in the inserted image clearly show that the plenty of bubbles are not located at the middle of Ag layers but rather closer to the V/Ag interface. The average diameters of the bubbles in the Ag and V layers are found to be approximately 1.5 and 1.0 nm, respectively.

Figure 2(b) shows the TEM image and the SRIM calculated concentration profile of He versus depth. The size and number of the bubbles in the depth of 0-60 nm are extraordinarily small. Our previous study on the He-irradiated pure bulk Ag and V under the same ion energy and fluence conditions showed that many bubbles were formed throughout the entire irradiated regions [13]. It is well known that He has negligible solubility in metals on the order of parts per billion or smaller [35]. In other words, if any measurable quantity of He atoms are introduced into metals, they will rapidly

aggregate into bubbles [29, 33, 35]. The above results of the V/Ag multilayer indicate that the interfaces in the V/Ag multilayer have high helium solubility, which delay the nucleation of He bubbles. This is due to the preferential trapping or absorbing of He atoms, bubbles, vacancies and He-vacancy complexes by the interfaces and their higher mobility at higher temperatures. Demkowicz et al.[27, 29, 36] have calculated the relative He trapping efficiency of many fcc–bcc interfaces which were in the K-S orientation relation. They found that misfit dislocation intersections (MDIs) in the interfaces of multilayers contained pockets of excess volume that readily trap He. He atoms trapped by the MDIs underwent a “platelet-to-bubble” transformation due to interface wetting. When the number of He atoms is below ~20, the He clusters grow as flat platelets which are too small to be seen by TEM. As the number of He atoms continuously increases, the He clusters present in the form of He bubbles, which can be observed by TEM. Therefore, the He concentration at which bubbles become detectable in multilayers is proportional to the areal densities of MDIs at the interfaces. It has been reported that V/Ag multilayers contained more than one type of interfaces [27, 28]. Our high resolution TEM observation shown in Fig. 1(b) also confirms that each successive layer in the multilayered V/Ag system is polycrystalline, and the neighboring Ag and V layers have more than one kind of orientation relations, suggesting a high density of MDIs in the V/Ag multilayers [29]. Therefore, the interface in the V/Ag multilayer has high He trapping efficiency, which increases the He solubility and delays the nucleation of He bubbles.

3.2. Retardation of bubble growth by interfaces

Figures 3(a)-(d) illustrate the in situ TEM observation of bubbles evolution in the V layers, the Ag layers and the V-Ag interfaces. Before annealing, the helium bubbles are observed to be circular white

dots that are mainly distributed in the Ag layers. As the annealing proceeds, the bubbles in the V layers grow slowly and maintain at approximately 1-2 nm in diameter. The bubbles in the Ag layers, however, grow rapidly and are limited by the layer thickness of ~6 nm. Bubbles nucleated at the interfaces grow into both V and Ag layers with a larger fraction of the bubble in the Ag layer. Bubbles that partially grow inside Ag layers are found to stop growing when they meet another interface. Bubbles marked in Fig. 3(d) indicate that the bubble growth is confined by the V/Ag interfaces. Oblong bubbles with lengths of greater than 6 nm (the sublayer thickness) are observed within the Ag layers along the direction parallel to the V/Ag interfaces. Figure 4 shows the bubble coarsening behavior as a function of annealing time and temperature, where the solid line displays the temperature ramping curve. It can be seen that bubbles 1-3 nucleate in the Ag layers, and grow rapidly to the size of the Ag thickness; bubbles 4 and 5 nucleate at the V/Ag interfaces and they grow into both Ag and V layers with a larger fraction of the bubble located in the Ag layer. Table 1 lists the bubble diameters in the Ag layers, the V layers and the entire multilayer film at different times. Bubble sizes in the entire multilayer are the weighted average of bubble sizes in V and Ag. The bubble size changes in the V and Ag layers as a function of annealing time are shown in Fig. 5. To obtain a reasonable statistics in bubble size determination, we counted approximately 200 bubbles from the same area of $\sim 250 \times 350 \text{ nm}^2$ of the TEM images. For the measurement of bubble size, in the case of spherical bubbles, the diameters of the bubbles were used as their sizes. In the case of non-spherical bubbles, we used the longest diagonals as their sizes. It can be seen that at the time mark of 4367 s, bubbles in the Ag layers increase rapidly to $\sim 4.5 \text{ nm}$ and continue to grow with a reduced growth rate. After a slower growth to $\sim 4.6 \text{ nm}$ at 4643 s, the bubbles appear not to grow any more despite the continued annealing of as long as additional $\sim 600 \text{ s}$. This growth behavior is consistent with the retardation effect of the bubbles by the interfaces, i.e.

bubbles in the Ag layer grow to the size of the layer thickness and then stop growing at the interfaces. A similar behavior of the bubble growth has also been observed in the Cu/Nb multilayers [25, 35]. At the same time, Figs. 3(a)-(d) and Fig. 5 show that bubbles in the V layers grow rather slowly from 1 nm at 0 s to 1.7 nm at 4125 s and appear to reach saturation in our available annealing time.

To interpret the difference of He bubble sizes in the V and Ag layers, it is important to understand how He atoms interact with solids at a nanometer scale. In fact, Goodhew et al.[37, 38] have studied the bubble growth mechanisms through extensive experiments and calculations. It has been found that He bubbles may grow through the following mechanisms [5, 37-44]: absorption of implanted He atoms; punching out of interstitials or dislocations loops; reabsorption of He atoms or vacancies emitted by small bubbles (Ostwald Ripening (OR)); migration and coalescence of bubbles (MC); absorption of vacancies. Different mechanisms may dominate the bubble growth process in different situations. Pertaining He irradiation, there are two common cases concerning the bubble evolution: irradiation at room temperature and followed by the subsequent annealing at higher temperatures; and irradiation at elevated temperatures [45, 46]. Our experiments focus on the first case. Therefore, the whole process of bubble growth will be discussed in two stages: the bubble formation (irradiation process) and the bubble growth process (annealing process) [5, 39].

During the He^+ ion irradiation, the supply of He atoms is provided and the lattice damage is sustained (target atom displacements). In addition, at a low irradiation temperature (e.g. room temperature), the migration of both thermal vacancies and irradiation-induced vacancies is slow. Therefore, the absorption of implanted He atoms and displacement-controlled athermal process such as formation of Frenkel pairs, dislocation loop punching, interstitial ejection contribute to the bubble formation [37, 39]. Wei et al. [3] studied the bubble formation in the He irradiated V/Ag multilayers,

and found that the bubble size in the Ag layer was much larger than that in the V layer. They [3] proposed that the higher threshold for dislocation loops punching and interstitial ejection in the V layers, as well as fewer clustered vacancies in the V layers result in the smaller sizes of bubbles in V. Therefore, for the bubble formation in the He ion irradiation process as shown in Fig. 2, bubbles in the Ag layers are larger than bubbles in the V layers.

For the bubble coarsening in the annealing process in our present work, the sample has been irradiated by He^+ ions with certain fluence at room temperature. Once the initial bubbles or He-Vacancy clusters are formed during the He irradiation at room temperature, further bubble coarsening and growth during the post-irradiation annealing occurs under the condition that there are no more He replenishment and added displacement damage, and the vacancy absorption is quick enough to prevent the athermal processes of dislocation loop punching and interstitial ejection [38, 39]. Therefore, the bubble coarsening and growth process are likely dominated by one or more of the remaining three mechanisms [5, 38, 43]: MC [41], OR [42] and vacancy absorption. The MC mechanism dominates at low temperatures (typically below $\sim 0.2 T/T_m$, where T_m is the melting temperature of the matrix). According to Goodhew's study [38], in Ni, the bubble growth rate induced by the MC mechanism is extremely lower compared with other mechanisms and can be neglected. The OR mechanism means that larger bubbles expand by absorbing He atoms emitted by smaller bubbles [42]. The driving force for this process is the reduction of the overall free energy of the system. Smaller size bubbles have higher surface chemical potential, which makes them unstable [47]. Large bubbles are more energetically favoured due to their lower specific surface area than smaller bubbles. Thus, He atoms on the surfaces of small bubbles tend to dissociate from the bubbles and migrate in the matrix. At the beginning of the annealing (Fig. 3(a)), all the bubbles are small and no significant differences

between bubble sizes exist, thus bubbles grow mainly by vacancy absorption. Irradiation can induce a high density of single vacancies and vacancy clusters in the matrix [44]. It has been reported by Trinkaus et al. [40, 48] that grain boundaries, interfaces and surfaces act as sources for vacancies, through vacancy absorption, thus small bubbles that are near the vacancy sources grow faster than those far away from the vacancy sources. Goodhew et al. [38] have reported that the vacancy absorption can induce fast bubble growth in FCC Ni and the growth rate was found to be as high as 10^5 nm/s. We believe that three factors are responsible for the rapid growth rate of bubbles in the Ag layers. First, the diffusion coefficient of vacancy relates to the homologous temperature [3], and the homologous temperature for vacancy migration is higher in Ag, leading to a higher diffusion rate of vacancies in the Ag layers. Second, the formation energy of vacancy in Ag is lower than that in V, leading to a higher density of vacancies in Ag [3]. Third, the interfaces and grain boundaries in the multilayer act as sources of vacancies [48]. Therefore, bubbles in Ag layers are easier to grow into larger sizes, and bubbles at the interfaces prefer to expand into Ag layers.

For the larger bubbles formed in the Ag layers, see Fig. 3(b), they may continue to grow by continuously absorbing vacancies or by absorbing He atoms dissociated from the smaller bubbles. It has been reported when the He irradiated FCC Ni was annealed at temperatures below $0.7 T_m$, bubbles grow mainly through vacancy absorption, and the overpressure of small bubbles is partially relaxed to moderate values, which results in the rapid bubble coarsening through the OR mechanism [48]. Golubov *et al.* [46] simulated the bubble coarsening at 600-900 °C, and concluded that the onset of bubble evolution takes place by vacancy collection, and small over-pressurized bubbles relax to equilibrium state. Therefore, at the beginning, bubbles both in V and Ag layers grow by vacancy absorption, and the further growth of larger bubbles in Ag layers occurs through vacancy absorption

and OR mechanism.

Besides the above mentioned reasons for the Ostwald ripening of He bubbles, following additional reasons may also make contributions. First, the pressure in small He bubble is very high. Thermal annealing leads to further increase of the pressure in small bubbles and may eventually lead to He emission/release. At higher annealing temperatures ($T > 0.5T_m$), the OR mechanism is expected to dominate the evolution of the He bubbles associated with accelerated dissolution of small bubbles. Second, the emission rate coefficient of He or vacancy from bubbles was size dependent, where smaller clusters have a higher tendency to emit a monomer and thus are more likely to shrink than larger clusters [49]. Third, it should be noted here that the existence of interfaces and other defects near the bubbles may affect the diffusion behavior of He [5].

For the retardation of He bubbles by interfaces, Hattar et al. [25] studied the confinement of He bubbles by the Cu-Nb interfaces. The mechanisms observed in the Cu-Nb interfaces should be also applicable to our V-Ag interfaces. In the irradiation and annealing processes, He bubbles grow through punching out dislocation loop and interstitial atoms, absorbing vacancies and He atoms. Bubbles in the Ag layers expand into the V layers must through one of the following ways: V atoms diffuse into Ag layers along the bubble surfaces; Ag vacancies diffuse into V layers; bubbles punch out V interstitials or interstitial dislocation loops into Ag layers. However, all these processes incurs an enthalpy penalty in face-centered cubic V. In other words, He bubbles in Ag are prevented from growing into V by the immiscibility of V in Ag [3, 25, 35]. Thus, bubbles in the Ag layers only grew to the thickness of the Ag layer and the further growth was restricted by the adjacent interfaces.

4. Conclusions

In situ annealing observation of the He irradiated V/Ag multilayer was conducted in TEM. The influence of interfaces on the bubble evolution was observed. Bubbles nucleated at the interfaces grow into both V and Ag sublayers with a larger fraction of the bubble in the Ag layer. Bubbles in the V layers grew slowly, while bubbles in the Ag layers grew rapidly and were eventually limited by the Ag layer thickness. Bubbles in V and Ag layers grew mainly by vacancy absorption and coarsening. The interfaces may retard the growth of bubbles. The trapping ability of the interfaces was described in the context of the interface microstructure that includes various crystalline orientation relations.

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CAPTIONS

FIG. 1. (a) Bright field cross-sectional TEM image and (b) HRTEM image of as-deposited V/Ag multilayer film, the selected area electron diffraction (SAED) pattern and the dark field cross-sectional TEM image of the multilayer are displayed in (a).

FIG. 2. (a) Cross-sectional TEM image of the as-irradiated V/Ag multilayer, the lower left corner of (a) is the magnification of the peak damaged region in the box, the SAED pattern and the calculated (SRIM) result for irradiation damage versus depth of the multilayer are displayed; (b) comparing the TEM micrograph of the He irradiated V/Ag multilayer (top) and the corresponding He concentration profile calculated by SRIM (bottom) reveals a critical depth at which no He bubbles are observed (indicated by dashed lines), even though the He concentration is not zero.

FIG. 3. (a)-(d) In-situ TEM images showing the evolution of the gas bubbles in the peak damage region of the multilayer under different annealing conditions, the defocus was -1100 nm.

FIG. 4. Bubble coarsening at different annealing stages, the curve shows the heating process.

FIG. 5. Bubble sizes evolution in Ag and V layers, respectively.

FIG. 1

FIG. 2

FIG. 3

FIG. 4

FIG. 5

TABLE(S)

TABLE 1. The evolution of the bubble diameters with standard deviation in the Ag layers, V layers and in the entire multilayer film.

Time (s)	0	1236	2218	3515	4125	4367	4643	5247
Temperature (°C)	25	269	426	430	542	542	542	542
Bubble size in Ag (nm)	1.5±0.1	1.6±0.3	1.9±0.6	2.4±0.7	3.8±1.5	4.5±1.7	4.6±1.3	4.6±1.3
Bubble size in V (nm)	1.0±0.1	1.1±0.1	1.3±0.2	1.5±0.2	1.7±0.3	1.7±0.31	1.8±0.3	1.8±0.3
Bubble size in V/Ag multilayer (nm)	1.4±0.2	1.5±0.3	1.9±0.6	2.2±0.8	2.7±1.4	3.1±1.9	3.2±1.4	3.2±1.4

Figure 1
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Figure 2
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