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Void formation close to stacking fault tetrahedra in heavily electron irradiated pure Ag and Cu

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Abstract

Pure Ag and Cu foils were irradiated with 1.25 MeV electrons at temperatures between 110 and 323 K in a high voltage electron microscope. For Ag foils irradiated at 300 K, SFT form an inhomogeneous lattice at first and then voids appear, the sites of which correspond to the SFT-lattice positions. Also at 270 K voids were found after a homogeneous formation of the SFT lattice, however, below 240 K voids were not observed. Void formation after SFT formation was also found for Cu specimens irradiated above 300 K. The nucleation process of voids is discussed in terms of the effect of a small amount of solute or gas atoms, which were segregated to SFT clusters under irradiation.

1. Introduction

It is well known that vacancy clusters exist in three types, viz., voids, stacking fault tetrahedra (SFT) and vacancy-type dislocation loops. If metals are irradiated with highly energetic particles such as electrons, ions, or neutrons, voids are generally formed at appropriate high temperatures. Macroscopic swelling due to the void formation is one of the major issues in irradiation damage of fusion reactor materials. However, the process of void nucleation, especially on the role of small amounts of gas or solute atoms in promoting void nucleation, is still not completely understood though a high amount of implanted He is known to obviously lead to bubble formation in a wider temperature range [1]. Vacancy clusters formed under irradiation at comparatively low temperatures are usually SFT or vacancy loops. From energy calculations void formation is not expected to occur in any of the high-purity f.c.c. metals [2], i.e., void formation at higher temperature must be have promoted by some extrinsic factor. It has been shown that the density of voids formed during neutron irradiation decreased strongly in the case where residual gas atoms were removed from Cu specimens, thereby suggesting that residual gas atoms play an important role on the void nucleation [3]. Moreover, a recent computer simulation using molecular dynamics shows that voids in pure copper can only nucleate upon introduction of gas atoms into small vacancy clusters [4].

The objective of the present work is to investigate the relation among the formation of three types of vacancy clusters in electron irradiated pure Ag and Cu, especially at lower irradiation temperatures where SFT or vacancy loops are usually formed.

2. Experimental

The wedge-shaped specimens used are from 99.9999% Ag and 99.999% Cu foils with a thickness of about 100 μm. After annealing to eliminate lattice defects, they were thinned by jet electropolishing. The electron irradiations were performed with 1.25 MeV electrons at temperatures between 110 and 323 K in a JEOL-ARM1250 high-voltage microscope equipped with a GATAN liquid-nitrogen-cooling stage and observations were done in situ. The irradiation flux was about 2×10^{24} e m^{-2} s^{-1} and the doses ranged from 10^{26} to 10^{28} e m^{-2}.

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3. Results

Fig. 1 shows the development of the defect structures in Ag irradiated along [0 0 1] at 300 K. Defect clusters with square dark contrast appear at first (Fig. 1(a)) especially in the upper part of the photos which is closer to the beam center. If projected along ⟨1 1 0⟩, the individual contrast features become triangular, indicating that they are SFT. No interstitial loops, which would rapidly increase in the size, were observed during the irradiation. SFT tend to nucleate next to the pre-formed ones along ⟨1 0 0⟩ axes, forming planer SFT walls. After growing to a critical size of about 6 nm, the square image of SFT often deformed as denoted by D and then the SFT collapsed to loops. (They did not grow rapidly thereby suggesting that they are vacancy loops.) Under prolonged irradiation, the number density of SFT decreased and voids appeared close to the SFT and increased in size, as shown by arrows.

A close relation between the formation of SFT and voids was found also for an irradiation along [0 1 1] at 300 K as shown in Fig. 2. Irradiation doses are (a) 2.1×10^{27} and (b) 4.1×10^{27} $e^- m^{-2}$, which, respectively, correspond to the time when SFT lattices have well developed and voids grown significantly. Defect structures were observed in two different imaging conditions, i.e., (a) adjusted to obtain strong contrast of SFT with a smaller deviation from Bragg reflection, and (b) with so-called void contrast far from any strong electron reflection. SFT are seen as triangles forming lattices in photo (a) and voids as bright images in photo (b). One should note that positional alignments of voids along [0 1 $\bar{1}$] are seen in photo (b), the direction of which corresponds to the projection of the (1 0 0) SFT lattice plane in photo (a). Moreover, direct evidence on the close relationship between the SFT and void formation are found in Fig. 3(a), which is a magnified view near the periphery of SFT lattice in a foil irradiated at 300 K, and in Fig. 3(b) which shows the isolated SFT in an Ag foil irradiated at 320 K. Voids are seen to nucleate at the sites of SFT, as shown by arrows in Fig. 3(a) and (b).

The formation of voids in Ag foils was investigated for irradiations at temperatures between 110 and 320 K up to a dose of about 8×10^{27} $e^- m^{-2}$ and the lowest temperature of the observation of voids was 270 K. Fig. 4 shows the defect structure developing from the electron beam center to the outer area at 270 K. The SFT lattice can be seen in the area far from the beam center while the contrast of the SFT lattice is becoming faint near the beam center. Voids with about 3 nm in diameter were found near the beam center as shown with bright circular images in the inset.

Also for Cu foils, a close relation between the void and SFT formation was found at 300 K and at 323 K. The lowest temperature of the observation of voids was 300 K for Cu.

4. Discussion

The present results show that void formation can occur in Ag and Cu specimens irradiated with electrons

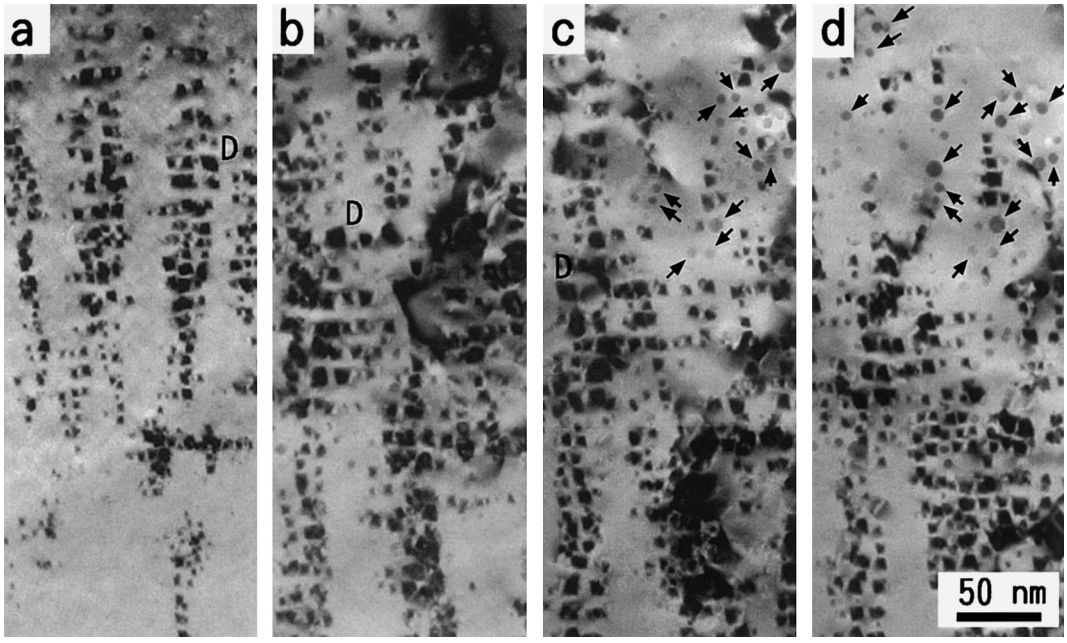

Fig. 1. Development of inhomogeneous SFT lattice structure and subsequent formation of voids in an Ag foil irradiated along [0 0 1] at 300 K: Doses were (a) 1.3×10^{27}, (b) 4.2×10^{27}, (c) 6.1×10^{27}, (d) 7.1×10^{27} $e^- m^{-2}$.

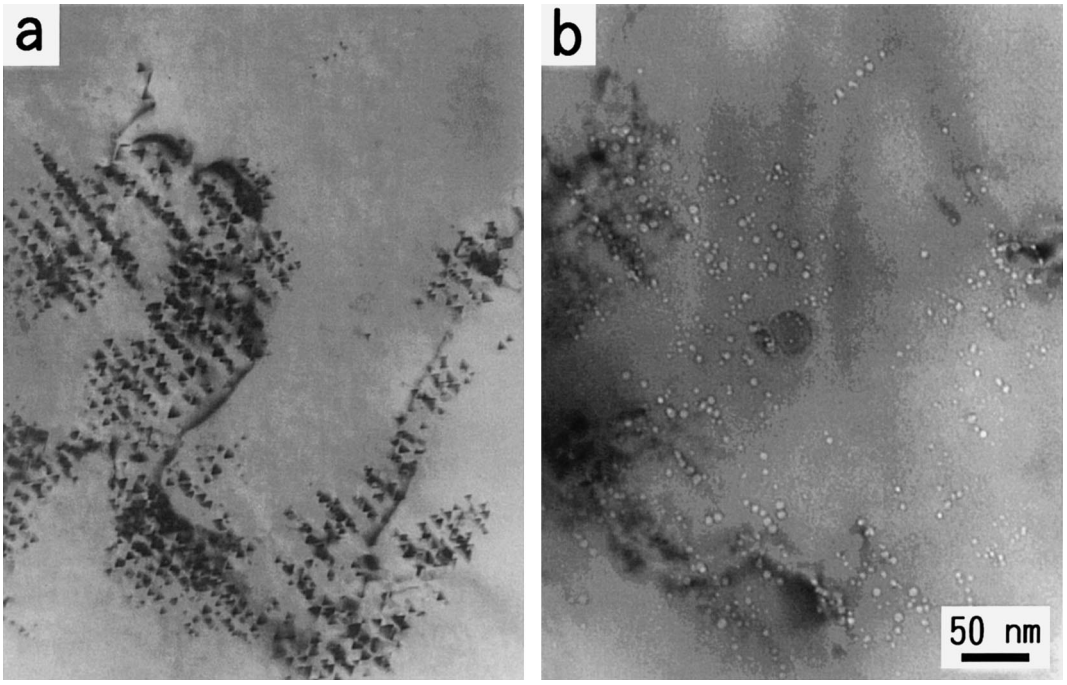

Fig. 2. Relation between SFT lattice formation and void distribution in the same area of an Ag foil irradiated along [0 1 1] at 300 K: (a) 2.1×10^{27} e^{-} m^{-2}; (b) 4.1×10^{27} e^{-} m^{-2}. Photo (a) is observed with a small deviation from the Bragg condition and photo (b) with a large deviation and objective underfocus.

at comparatively low temperatures, if sufficiently high doses are applied. From a practical standpoint, the void swelling will not be of concern for Ag and Cu exposed in the present irradiation temperature range (below 323 K) as the number density is low and the size is small. However, the present results are of interest for understanding the nucleation process of vacancy clusters, as the three types of vacancy clusters, SFT, vacancy loops and voids, appeared not at the same time under irradiation.

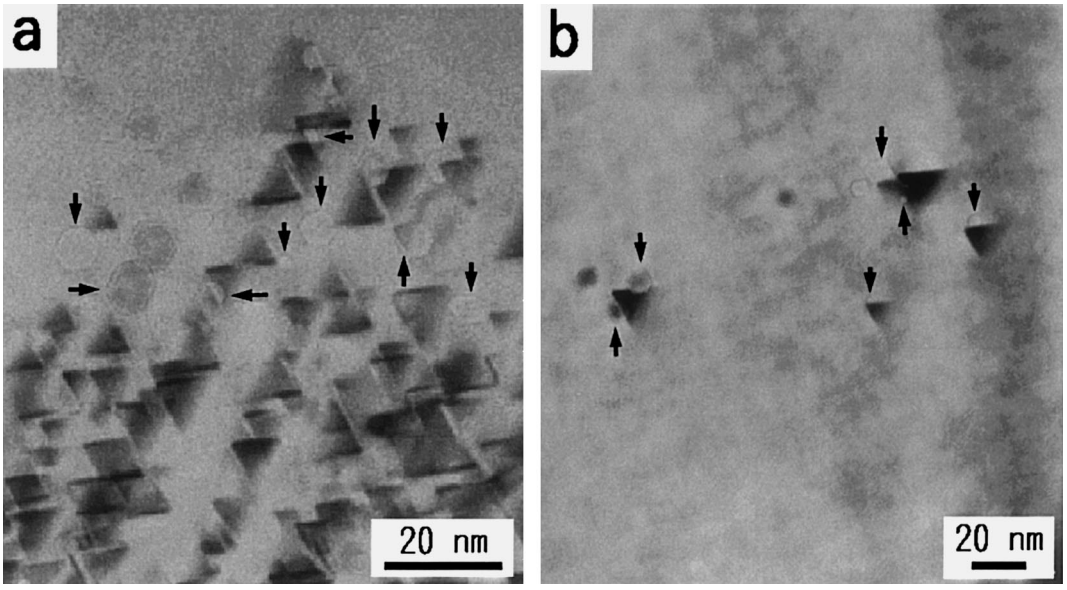

Fig. 3. Preferable nucleation of voids at the sites of SFT in Ag foils: (a) 300 K, 4.0×10^{27} e^{-} m^{-2}; (b) 320 K, 2.5×10^{27} e^{-} m^{-2}.

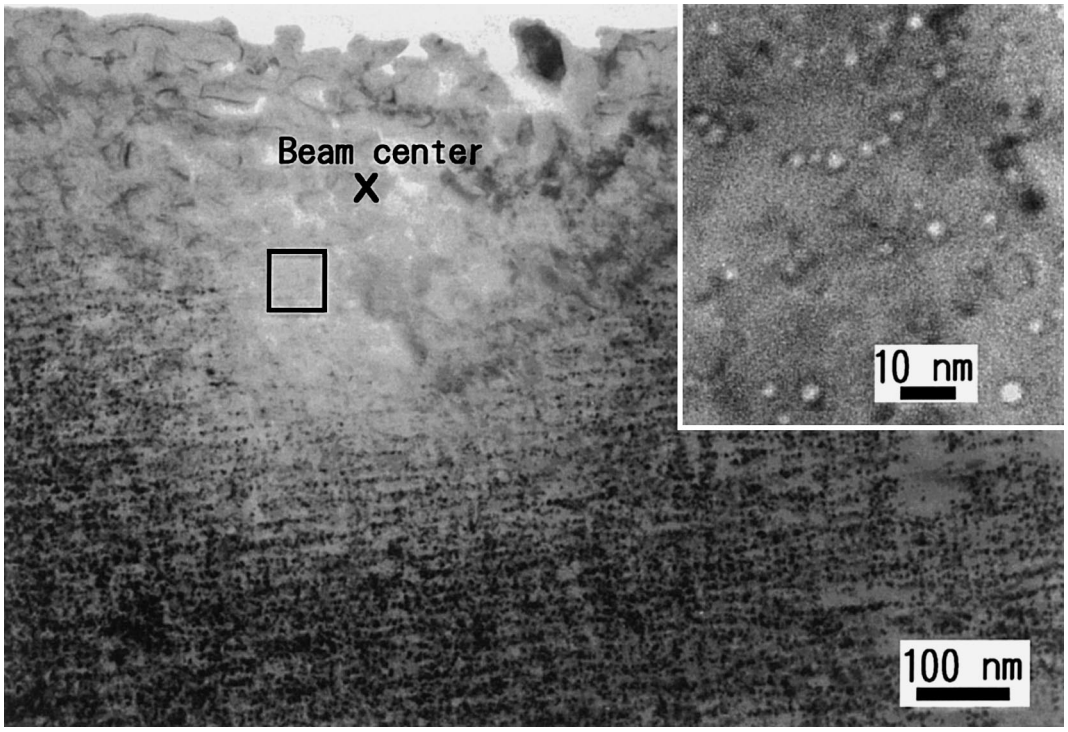

Fig. 4. Formation of homogeneous SFT lattice structure and voids in an Ag foil irradiated along [0 0 1] at 270 K to a dose of 7.1×10^{27} e^{-} m^{-2}. Inset is a magnified view of the voids in a region denoted by a black square.

For Ag irradiated at 300 K, the first observable defect clusters were SFT and no interstitial loops were found. The evolution of SFT formation leads to a well arranged lattice of CsCl structures which has been systematically investigated [5,6]. From these analyses, the irradiation temperature of 300 K corresponds to a regime, where inhomogeneous SFT lattice formation occurs. In this regime, SFT tend to nucleate in the vicinity of pre-existing defects, i.e. new SFT appear next to preformed ones along ⟨1 0 0⟩ axes, thus forming planer SFT walls. The development of SFT in the present study coincides with these results and the present new finding of void formation corresponds to the late stage of SFT lattice formation. The lowest temperature of the void formation for Ag, judged by the present TEM observation, was 270 K as shown in Fig. 4. This temperature almost corresponds to the critical temperature (280 K), where thermal drift of vacancies starts to affect the formation of the SFT lattice structure [6]. For copper, on the other hand, the lowest temperature of the observation of voids is 300 K, which also corresponds to the transition temperature (290 K). Both critical temperatures are about 0.2 T_m where T_m is the melting temperature. These results suggest that thermal drift of vacancies is necessary for the void formation.

The present finding of void formation after SFT formation has been scarcely reported for pure metals as far as we know, though the transformation from SFT to vacancy loops has been observed so far [7,8]. This is probably because it is rather difficult to detect very small size of voids without high resolution electron microscope, as we could not find the void formation with an old type of high voltage electron microscope (AEI EM7) with the same purity of specimens and similar irradiation condition [5]. The detailed observation of the positional relation between SFT and voids, especially for isolated SFT formed in Ag irradiated at 320 K (Fig. 3(b)) shows that voids nucleate at the sites of SFT. A similar close relation between SFT and voids has been reported by Kojima et al., for the austenitic Fe–13Cr–14Ni alloy irradiated with 1 MeV electrons at temperatures between room temperature and 659 K [9]. They have found conversion of SFT to voids in the temperature range at which both SFT and voids were coexistent, and considered that local effects of the dilatation field at the corner of SFT and the segregation of solute atoms enhance the void nucleation. A similar explanation on the nucleation of voids found in the present study may be possible though we have utilized pure metals. A drastic decrease of the number density of voids has been observed in neutron irradiated pure Cu due to the removal of residual gas atoms. Thus a small amount of gas or solute atoms in pure metals may affect void nucleation [3]. The agglomeration of point defects

at sink sites such as defect clusters, dislocations or grain boundaries would condense a small amount of gas or solute atoms and may lead to the void nucleation. Therefore, the present new finding of the formation of voids at the sites of SFT in electron irradiated pure metals is considered to be an indirect evidence of the necessity of a small amount of gas or solute atoms [4] on void nucleation.

5. Conclusions

In order to investigate the nucleation process of voids, we have irradiated pure Ag and Cu with 1.25 MeV electrons at temperatures between 110 and 323 K. The lowest temperature of the void formation was 270 and 300 K for Ag and Cu, respectively, i.e. about 0.2 T_m. The void formation was observed after the formation of SFT lattice, closely relating to the sites of SFT. The nucleation of voids close to SFT in these pure metals was considered to be induced by accumulation of a small amount of residual solute or gas atoms in the pure metals to SFT under long term irradiation, supporting the idea of necessity of a small amount of solute or gas atoms on the void nucleation.

References

[1] K. Niwase, T. Ezawa, T. Tanabe, F.E. Fujita, J. Nucl. Mater. 160 (1988) 229.
[2] S.J. Zinkle, L.E. Seitzman, W.G. Wolfer, Philos. Mag. A 55 (1987) 111.
[3] Y. Shimomura, I. Mukouda, M. Kiritani, S. Kojima, F.A. Garner, J. Nucl. Mater. 212–215 (1994) 352.
[4] Y. Shimomura, M.W. Guinan, T. Diaz de la Rubia, J. Nucl. Mater. 205 (1993) 374.
[5] N.Y. Jin, F. Phillipp, A. Seeger, Phys. Stat. Sol. (a) 116 (1989) 91.
[6] A. Seeger, N.Y. Jin, F. Phillipp, M. Zaiser, Ultramicroscopy 39 (1991) 342–354.
[7] M.O. Ruault, Philos. Mag. 36 (1977) 835.
[8] H. Fujita, T. Sakata, H. Fukuyo, J. Appl. Phys. 21 (1982) L235.
[9] S. Kojima, Y. Sano, T. Yoshiie, N. Yoshida, M. Kiritani, J. Nucl. Mater. 141–143 (1986) 763.