Self-ion irradiation has been used to study the collapse of displacement cascades in pure copper, and molybdenum in the temperature range 20°C - 500°C. In 30 KeV self-ion irradiation of copper the number of vacancy loops formed from the cascades was observed to remain constant at temperatures below 300°C but to decrease sharply at temperatures above this. In 60 KeV self-ion irradiation of molybdenum a gradual decrease in the number of vacancy loops was observed above 200°C, an order of magnitude decrease occurring by 500°C. The variation in numbers is compared with the predictions of a model which considers only thermal vacancy emission from the loops. This was found to give very good agreement for the copper results but not for the molybdenum results. Preliminary results are also reported on the effect of temperature on the Burgers vectors of the vacancy loops and on 60 KeV self-ion irradiation of nickel.
EXPERIMENTAL PROCEDURE

The experiments were carried out on specimens cut from single crystals, the specimen normals being \(<110>\) and \(<123>\) for copper, \(<110>\) for molybdenum and close to \(<110>\) for nickel. The techniques used in preparing the copper and molybdenum crystals and their purity has been discussed elsewhere\(^1\). The nickel crystal, obtained from Metals Research Ltd. was 4N pure. Thinned foils of each material were irradiated in either the Harwell Heavy Ion Accelerator or in the Harwell Lintott Separator. In the former the error limits on quoted temperatures were \(\pm 25^\circ\text{C}\) and in the latter \(\pm 5^\circ\text{C}\).

Following irradiation at room temperature, specimens of all three materials contained a uniform distribution of vacancy defects and the principal objective of the electron microscope studies was to determine the influence of irradiation temperature on defect geometry, number density, \(N_d\), and size distribution. Two methods were used in determining the Burgers vectors of loops. The first was the so-called 1 analysis involving measurements of the directions of black-white streaking of dynamical images from small loops\(^2\). This method is known to be valid for Frank loops in copper but breaks down when applied to perfect loops particularly in more isotropic cubic crystals such as molybdenum. The second and more positive method, which we used in particular for molybdenum, was to compare the symmetry of dynamical black-white images obtained under well-defined two beam diffracting conditions with theoretically simulated images from known loops in an isotropic cubic crystal\(^3\).

RESULTS

Considering first the results from copper irradiated with 30 KeV self-ions to a dose of \(4 \times 10^{12}\) ions cm\(^{-2}\). Fig. 1 shows how \(N_d\) varies with irradiation temperature in \(<123>\) foils in the range 20–425\(^0\text{C}\). The defect counts were carried out on micrographs obtained with \(g = (200), \omega = 0\) at a foil normal \(2 = [025]\). It can be seen that \(N_d\) remains constant up to 300\(^0\text{C}\) and then decreases by about an order of magnitude between 300 and
The image diameters were measured on large micrographs obtained with $g = (111)$, $w <1$, and $z = <110>$. Fig. 2 shows the image size distributions based on measurements of 300 defects for specimens irradiated at room temperature and $350^\circ C$. A shift in the distribution towards larger image sizes occurs in the $350^\circ C$ foils. This is reflected in the average image diameter which increases from $46 \%$ at $20^\circ C$ to $55 \%$ at $350^\circ C$.

A preliminary assessment of the defect geometry in $<123>$ foils was carried out by applying the $\frac{1}{2}$ analysis to dynamical black-white images on micrographs obtained with $g = (111)$ and $g = (220)$. The results showed that in the specimens irradiated at room temperature $\frac{1}{2}$ for $97\%$ of the loops lies along one of the $<111>$ directions but as the irradiation temperature increased an increasing fraction of the images had $\frac{1}{2}$ along $<110>$ or some direction between $<111>$ and $<110>$. Because of uncertainties in the interpretation of results from a simple $\frac{1}{2}$ analysis, more detailed studies are being made of defects in $<110>$ and $<123>$ foils by combining measurements of $\frac{1}{2}$ using different
diffraction conditions as defined by $g$ and $z$-with an examination of image symmetry. The results of such an analysis on $<110>$ and $<123>$ foils irradiated at 350°C have led to conclusions which differ in two respects to the indications from the preliminary survey. Firstly, as shown in Fig. 3, stacking fault tetrahedra and dissociated Frank loops can be identified in foils of both orientations. Secondly, the more comprehensive analysis did not show any evidence for perfect loops being present in foils irradiated at 350°C.

Fig. 3. Micrographs of foils irradiated at 350°C with $6 \times 10^{17}$ 30 KeV Cu$^+$ ions/cm$^2$, (a) $<011>$, (b) $<123>$.

Fig. 4. Micrographs of foils irradiated at 350°C with $7.4 \times 10^{17}$ 60 KeV Mo$^+$ ions/cm$^2$, (a) $g = (2\overline{1}1)$, (b) $g = (2\overline{1}7)$. 
Turning now to the results from the irradiation of molybdenum with 60 KeV Mo⁺ ions to a dose of $7.4 \times 10^{11}$ ions cm⁻² in the temperature region 20°C - 535°C. The variation in $N_d$ with irradiation temperature is included in Fig. 1 and it can be seen that in contrast to copper, $N_d$ starts to fall between 150 and 200°C and has decreased by about an order of magnitude at 450°C. The Burgers vectors of loops were analysed using micrographs obtained with $\mathbf{g} = \frac{1}{4} (2\bar{1}1)$ or $\mathbf{g} = \frac{1}{4} (21\bar{7})$ at $\mathbf{g}$ close to $<011>$ and matching the symmetry of black-white images with that of theoretically computed images from known edge loops. In the foils irradiated at high temperatures, $>400°C$, a good correspondence was obtained between a large proportion of the observed images and computed images for the two sets of perfect edge loops having $\mathbf{b}$ lying in the foil plane, i.e. $\mathbf{b} = \frac{1}{2} [\bar{1}1\bar{7}]$ or $\mathbf{b} = \frac{1}{2} [11\bar{7}]$. In these cases $|\mathbf{g} \cdot \mathbf{b}| = 0$ or 2 for the two $\mathbf{g}$'s used and the images are easily identified as shown by the examples labelled A and B in Fig. 4. However, a number of the images are distorted from these ideal configurations (examples are labelled C in Fig. 4). Applying the results from calculation by Haussermann et al., we deduce that these distorted images are from non-edge perfect loops. Examination of the occurrence of such distorted images as a function if irradiation temperature has led to the general conclusion that the loops from initially on $\{110\}$ but shear to a perfect configuration at an early stage. However, they can only rotate to a pure edge configuration during irradiation at elevated temperatures. It is emphasised that we have obtained no evidence for loops having $\mathbf{b}$ other than $\frac{a}{2} <111>$.

The image size distribution for specimens irradiated at room temperature and 535°C is shown in Fig. 2 and this was obtained from measurements of 200 defects imaged with $\mathbf{g} = (01\bar{7})$, $\mathbf{w} <\bar{1}$ at a foil normal of $<155>$. The average diameter increases from 27.7 $\AA$ at 20°C to 41.5 $\AA$ at 535°C.

Lastly, preliminary measurements have been made of the defect density in nickel irradiated with 60 KeV Ni⁺ to a dose of $6 \times 10^{11}$ ions cm⁻² at temperatures between 20°C and 475°C. The results show that there is no significant decrease in $N_d$ below 300°C but defects were not observed in specimens irradiated at 475°C. It is also interesting to note that some dissociation of the Frank loops towards stacking fault tetrahedra was
observed in foils irradiated at 200°C.

DISCUSSION

The results have exposed two important aspects of how temperature influences cascade collapse in fcc and bcc metals. The first is the fundamental difference that exists between copper and molybdenum regarding the temperature dependence of vacancy loop survival. The second is the effect irradiation temperature has on defect geometry.

Considering first the decrease in \(N_d\) with increasing irradiation temperature, the copper results can be accounted for by a simple thermal emission model. In this model it is assumed that cascade collapse to form loops is athermal and thus we can define a loop generation rate \(K_L\) which is simply the product of dose rate and defect yield, i.e., the number of loops formed per incident ion. The defect yield in self-ion irradiated copper at room temperature is \(0.35\). This generation rate is then balanced against the loss rate of loops due to shrinkage by thermal vacancy emission. The rate equation for this latter process is:

\[
\frac{dN}{dt} = -\frac{2}{b} D_S \exp \frac{(\gamma + F_{el})}{kT} \frac{B^2}{b^2} \tag{1}
\]

where \(D_S\) is the self-diffusion coefficient, \(\gamma\) is the stacking fault energy of faulted loops, \(F_{el}\) is the elastic self-energy of the loop and \(\frac{1}{b^2}\) is the cross-sectional area of a vacancy. The experimentally measured values of \(N_d\) represent the number of visible loops, i.e., those having a radius greater than the minimum visible radius, \(r_{min} = 10 - 15 \, \AA\). Thus, we can calculate the time \(t_L\), taken for the loops to shrink from an initial mean radius \(r_0\) (determined from the room temperature irradiated specimens) to \(r_{min}\) by integrating Eq. (1), i.e.

\[
\frac{t_L}{b} = \frac{B^2}{2D_S} \int_{r_0}^{r_{min}} \exp \left(\frac{(\gamma + F_{el})}{kT} \frac{B^2}{b^2}\right) dr \tag{2}
\]

Then the calculated loop numbers at any temperature \(T\) is given by

\[(N_d)_T = K_L(t_L)_T\]  

We show in Fig. 1 the calculated variation in \(N_d\) with
irradiation temperature for copper and the striking feature is the close agreement with the experimental results. Although the $N_\theta$ versus temperature data from nickel is incomplete, it would appear again that they can be accounted for by a thermal emission model.

With regard to the results from molybdenum, it is clear that the decrease in $N_\theta$ at temperatures as low as $200^\circ C$ cannot be accounted for by a thermal emission model. It is emphasized that the form of the $N_\theta$ versus temperature curve is similar to that obtained by Bentley\textsuperscript{6} from neutron irradiated molybdenum. We can therefore rule out any explanation that involves the close proximity of a free surface. A particularly important feature of Bentley's results is that the vacancy loop population is replaced by a fine distribution of voids. Moreover, in the present experiments we observed that the mean loop radius increases with increasing irradiation temperatures. Thus, we conclude that the reduction in $N_\theta$ is not due to increased recombination of interstitials with vacancies in the cascade. Rather, we believe that the observed behavior is a consequence of a temperature dependence in the cascade collapse process itself in bcc metals.

An important factor in this may be the extremely high stacking fault energy characteristic of molybdenum and other bcc metals, so that when the vacancy migration temperature has been achieved the cascades break up rather than collapse, thus releasing the vacancies for void formation. Clearly this fundamentally different behavior in bcc metals is of considerable importance and needs to be explored more thoroughly.

The question of the effect of irradiation temperature on the loop geometry is obviously quite complicated. However, a fundamental difference has been seen between the behavior of loops in copper and molybdenum. In the bcc molybdenum the loops unfault at room temperature and the effect of increasing the irradiation temperature is to cause the loops to rotate to the pure edge configuration. In copper the loops remain faulted and at elevated temperature the dissociation of the Frank loops can result in complete stacking fault tetrahedra, in agreement with the work of Schindler\textsuperscript{7}. The results of the irradiation at $350^\circ C$ suggest that the surface influences the dissociation of the Frank loops in copper. In molybdenum the complete analysis, to be reported elsewhere, suggest that the surface influences
which particular perfect loop the \( \{110\} \) loops unfault to.

REFERENCES


7. R. Schindler and M. Wilkens, unpublished work.