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ABSTRACT

14 MeV neutron damage has been investigated in gold by means of transmission electron microscopy (TEM). The sites of energetic displacement cascades are visible due to the presence of vacancy clusters formed by the collapse or rearrangement of vacancies within the depleted zones. We find in the 14 MeV n induced cascades a strong tendency towards subcascade formation. On the average 1.8 clusters are formed per cascade. Individual cascades with as many as 6 subcascades have been found. The number densities of clusters as well as the number densities of cascades are proportional to the fluence. The cross section for the formation of visible cascades is $\sigma_c = 3.3 \times 10^{-24} \text{ cm}^2$. It can be shown that recoils from elastic neutron scattering events can account for only about 20% of the visible cascades. The cross section corresponding to the balance of the observed cascades is, within experimental error, equal to the nonelastic neutron scattering cross section. This indicates that all non-elastic scattering events lead to the formation of a visible cascade. We find quantitative agreement with what is expected from heavy ion bombardments regarding the cross sections involved, however, estimates of the average cascade energy in the 14 MeV neutron bombardments are somewhat higher than expected.

*This work was performed under the auspices of the U. S. Atomic Energy Commission.

INTRODUCTION

Recoil atoms

The D-T fusion reactor components will be subject to irradiation by 14 MeV neutrons. Such energetic neutrons will be able to transfer considerable amounts of kinetic energy to the lattice atoms at which they are being scattered. The energies imparted to the primary recoil atoms range up to hundreds of keV even for relatively heavy nuclei. In the present paper we study the displacement cascades that are initiated by such energetic primaries.

Observation of displacement cascades by means of TEM

Theoretical models of cascades at relatively low energies (< 50 keV) have predicted that the cascade contains a central vacancy rich region (depleted zone) while the corresponding interstitials reside more at the periphery in the form of small clusters and, at low temperature, also as single interstitials. The excess vacancies in the depleted zone can collapse into a dislocation loop or rearrange into more complex structures. These vacancy clusters have indeed been observed in Cu and Au by means of Transmission electron microscopy (TEM).¹⁾ The interstitials and interstitial clusters are usually not visible when TEM is used, but the interstitials have been observed by field ion microscopy (FIM).²⁾ In our present work we use TEM on gold irradiated with 14 MeV neutrons at doses where the cascades are not expected to overlap significantly. We, therefore, essentially study the vacancy clusters that are directly formed in individual cascades at the site of the depleted zone.³⁾ The reason for choosing gold as a model substance lies in the fact that it has been well established that all cascades produce clusters visible by TEM provided the cascade receives energy in excess of 40 to 50 keV.⁴⁻⁷⁾ In lighter metals, as for example in Cu and Al, the defect density

in the cascade is lower and the clusters are smaller than in Au, to the extent that the cascade clusters are observed by TEM with only a low probability (Cu)^{5,8)} or not at all (Al). For those metals TEM is not very suited to study details of the primary cascade defect structures because of the limited resolution. On the other hand, the basic cascade structures are expected to exhibit strong similarities. Therefore, our results should be applicable to other metals if proper scaling factors are introduced. Although TEM gives only a rough picture of the defect structures in a cascade, this method is ideally suited to look at very energetic cascades, while computer simulations as well as FIM studies become difficult to manage at energies greater than a few tens of keV. TEM also has the advantage that it is easy to examine a larger number of cascades which is necessary because of the statistical nature of the cascade.

Heavy ion induced cascades

A large body of information on cascades in Au is available from heavy ion bombardments and in particular from self ion bombardments^{4-7,9,10)}. We shall draw on those results for a comparison with the cascade structures observed under 14 MeV neutron bombardment. The most important aspects of the heavy ion results are the following:

1. The probability of forming a cluster is known as a function of the cascade energy.
2. Splitting into subcascades occurs at high cascade energies (> 50 keV).
3. The amount of splitting is related to the average cascade energy.
4. The separation of vacancies and interstitials takes place with rather high efficiency.
5. The number of clustered vacancies per cascade is roughly proportional to the energy available for making nuclear displacements.

Figures 1 and 2 show examples of self ion induced displacement cascades. In Fig. 1 cascades of 50 keV energy are shown. At this energy every cascade produces a visible vacancy cluster. Figure 2 depicts a gold specimen irradiated with an extremely low dose of 200 keV Au^+ ions. The splitting into subcascades is quite apparent. We shall see that similar cascades are also produced under 14 MeV neutron bombardment.

EXPERIMENTAL

Specimen preparation

Au-films of (001) orientation were grown epitaxially on cleavage surfaces of rock salt. The Au films were annealed in air at 350°C for 30 min. and mounted on Cu and Au microscope specimen grids. The film thickness was 810 Å as determined by gravimetry. Six films were mounted in an Aluminum holder. Each film was positioned between two small gold rings and gold discs. In this way the Au-films were almost completely surrounded by Au surfaces. This was done in order to have the same recoil density throughout the thickness of the specimen. This also minimizes the effects that might come from recoil atoms other than Au, that would be expected to impinge from outside onto the surfaces of the specimen. The Al holder was sealed inside a copper capsule. The capsule was evacuated and backfilled with approximately 1/10 atm. of helium. The positions of the specimen are schematically indicated in Fig. 3.

14 MeV neutron irradiation

The capsule was irradiated at room temperature with 14 MeV neutrons at the Lawrence Livermore Laboratory rotating target neutron source to a nominal fluence of 10^{16} n/cm^2 . At the position of the capsule the neutron flux dropped off rather strongly with distance from the source. Therefore, in order to be able

to determine the neutron fluence at the specimens, Nb dosimetry foils were attached to the outside of the capsule at 6 points. The positions of the dosimetry foils are indicated in Fig. 3. The average fluence at each foil was determined from data obtained from γ -ray counting and a proton recoil telescope. R. A. Van Konyenburg has described the details of the method in ref.11. The fluences that were obtained from 6 dosimetry foils at the capsule closely showed an inverse square dependence with distance from the center of the source. The fluence values at the center of each specimen were, therefore, obtained from this dose-distance relationship. The fluence values obtained in this way ranged from 7×10^{15} to 2×10^{16} neutrons/cm².

TEM examination

After irradiation the specimens were kept at ambient temperature for several weeks before they were taken out of the capsule and examined in transmission in a Siemens Elmiskop 1a. The electron optical magnification was 70,000X. Evaluation of the data was made from prints at 280,000X. The electron micrographs were taken in bright field within about 5° of the [001] direction, usually in the vicinity of [1,1,16] under "kinematical" conditions. We found that rather sharp and well defined black spot images could thus be obtained near [001] under conditions where many beams were excited rather symmetrically to the row of systematic reflections. Similar observations have also been made by M. Rühle¹²⁾ Stereoelectron micrographs were taken by symmetrical tilts relative to the plane of the film.

RESULTS

Cascade and subcascade clusters

Figure 4 shows a stereo pair of gold film that has been irradiated to a fluence of 7×10^{15} n/cm². One can clearly recognize defect clusters in the form

of the black spots. It can be seen that the clusters are distributed throughout the thickness of the foil. From the heavy ion results we infer that the clusters are of vacancy type and represent to a large part Frank dislocation loops. The bunching of clusters is quite apparent. Each group of closely spaced clusters is produced in an individual cascade. At this low fluence the individual cascades are well separated in space and the probability for overlap is quite low. The stereoscopic view also gives quite a clear impression of the group of spots that belong together. This bunching of spots is indicative of subcascade formation. Subcascade formation has first been observed in fission fragment irradiations¹³⁾ and self ion irradiations have indicated that rather strong splitting into subcascades takes place in the region above 100 keV.^{9,10)}

Fluence dependence of the number density of clusters

Let us now consider the total number of defect clusters, irrespective of whether more than one of these are produced per cascade. In any case, the number of clusters will be proportional to the fluence if the clusters are directly produced in cascades and as long as no overlap between cascades occurs. The number densities of the vacancy clusters as a function of dose are shown in Fig. 5. We see that the data fall reasonably close to a straight line through the origin. The vertical error bars correspond only to the error due to counting statistics, while the horizontal error bars only take a $\pm .5$ mm uncertainty in the specimen position into account. The latter then produced an additional neutron fluence uncertainty. The absolute accuracy of the neutron dosimetry at the positions of the monitor foils is estimated to be $\pm 7.5\%$. We estimate that the total absolute error in the slope of the straight line in Fig. 3 is on the order of $\pm 20\%$. The slope in Fig. 3 directly gives the average number of defect clusters per cm along the path of a neutron. We find

$\frac{N}{\phi} = .34$ clusters per cm. We shall not pursue this any further for the moment, because we know that the individual clusters are not produced randomly along the path of the neutron. This follows from the fact that the clusters are bunched at the sites of energetic cascades as seen in Fig. 4 for example. To check this further we have made a χ^2 test of our counting data. The number of clusters per unit area is usually determined by overlaying the photographs with a grid consisting of squares of $2.5 \times 10^{-11} \text{ cm}^2$ area and counting the number of spots in each grid square. If the individual black spots were distributed randomly, the population of the squares should follow a Poisson distribution. We find very strong deviations from a Poisson distribution which confirms what seems quite obvious from a direct inspection of the photos.

Cluster size distribution

The size of the individual black spot defects relates to the number of vacancies that have clustered within the depleted zone of a subcascade or cascade. We shall see later that the size distributions can be used to make an estimate of the damage production efficiency in cascades. The diameter of the clusters was measured for specimen #1. The results are shown in Fig. 5a where the relative population of 10 Å size groups is depicted. The average diameter turned out to be $\bar{d} = 48 \text{ Å}$, while $(\bar{d}^2)^{1/2} = 51 \text{ Å}$. Although the distribution is peaked at values clearly above the resolution limit of the electron microscope it is very likely that the visibility limit is to a large part responsible for the drop off towards the lower sizes. The size distribution is quite similar to the distributions that we find in Au-self ion bombardments in the energy region where the cascades split into subcascades. In self ion bombardments the size distributions hardly change above a cascade energy of 100 keV. This is a direct consequence of the splitting into subcascades. For

comparison we show in Fig. 5b the size distribution observed for 250 keV cascades. In this case the average diameter is also $48 \text{ \AA}$. By comparing Figs. 5a and b we see that the size distribution is quite similar to the 14 MeV neutron case. We should like to emphasize that these size distributions represent the sizes of the subcascade clusters and are, therefore, not directly related to the total size of the cascade or subcascade. However, their size obviously depends on the number of point defects in the clusters.

Cascade cross section

So far we have mainly looked at the total number and sizes of subcascade clusters. If we now count the total number of visible cascades we can relate this to the number of energetic recoils that are produced during the 14 MeV neutron irradiation. For determining the number density of cascades we used the following procedure: Each bunch of spots is counted as one cascade if all of the spots within the bunch fit into a circular area of a given diameter. If several clusters are close-by, but cannot be fitted within one circle, we assume that more than one cascade is present and we determine the number of cascades from the minimum number of translations of the circle that are necessary to cover all of the defect clusters. This procedure gives the correct number of cascades if the density of cascades is so low that the probability of overlap between the circles is low and if even the most energetic cascades can be fitted within the standard circle. The average transverse diameter of the most energetic cascade that can be produced in an elastic collision with a 14 MeV neutron (280 keV) is about $300 \text{ \AA}$ as determined from our heavy ion results. Therefore, a $400 \text{ \AA}$ diameter should just be sufficient to accommodate almost all of the most energetic cascades. On the other hand, the area is small enough to give an overlap error of only about 2% of our lowest fluence.

Specimens number 1 and 6 were counted in this way. For the higher fluences we reduced the diameter of the test circle to $300 \text{ \AA}$, because the overlap error would have become too large. On the other hand, we know from the self ion irradiations that a $300 \text{ \AA}$ diameter circle will not accommodate the most energetic cascades. A test count with the $300 \text{ \AA}$ circle at the low dose showed indeed a 7% higher cascade count compared to the count obtained when using the larger test circle. We felt this was small enough to also try to determine the number density of cascades at the higher doses. Figure 7 shows the results. We see that the number density of cascades is proportional to the fluence. From the slope of the straight line $\frac{N_c}{\phi}$ in Fig. 5 we now determine the cross section for the formation of visible cascades. It turns out that $\sigma_c = \frac{N_c}{N\phi} = (3.3 \pm .7) \times 10^{-24} \text{ cm}^2$, where N is the atomic density. The mean free path before making an energetic neutron collision leading to a visible cluster is $\lambda = \frac{\phi}{N_c} = 5.2 \text{ cm}$. Using the cluster density determined above we can now determine the average number of clusters per cascade: $\bar{N}_s = \frac{N_s}{N_c} = 1.8$.

Cascade clustering efficiency

As mentioned above, the size of the clusters can give an indication of the number of vacancies that they contain. If we assume that the clusters consist of circular Frank dislocation loops on (111) planes, we get an estimate of the average number of clustered vacancies per cascade from the size distribution and the number density of subcascade clusters. We find $\bar{N}_v = 340$ vacancies per average subcluster. In heavy ion irradiations we have observed a rather high clustering efficiency, when we compare the number of clustered vacancies with the total number of defects predicted by simple theories.¹⁴⁾ We can calculate the total number of Frenkel pairs N_F that are produced in a cascade using a simple modified Kinchin and Pease expression:¹⁵⁾

$$N_F = \frac{E_D}{2E_d}$$

where E_D is the damage energy (electronic losses subtracted from the cascade energy) and E_d is the threshold energy for displacement. The average number of clustered vacancies per cascade is $N_V = \frac{N_V}{N_C} \cdot \frac{N_S}{N_C}$. It is convenient in this connection to use a cascade clustering efficiency factor $\xi = \frac{N_V}{N_F}$. From the self ion irradiations we found that ξ is remarkably constant for cascade energies above 50 keV and we obtain $\xi \sim 0.3$. Assuming the same clustering efficiency we can now estimate the average cascade energy in the 14 MeV neutron irradiation. We find $E_D \sim 135$ keV, which seems rather high, particularly if one considers that this implies an average total cascade energy of 190 keV if electronic losses are taken into account.

We can also try to estimate from the number of vacancies in cascades the energy that has been expended in individual cascade events. Such a determination of the cascade energy is rather difficult, because of the statistical nature of the cascades, which causes large fluctuations in the cascade structure and number of defects. Our self ion irradiations have shown this relatively large spread in the number of vacancies clustered in monoenergetic cascades. However, we have never observed numbers which exceed the theoretical modified Kinchin and Pease value ($\xi = 1$). This immediately gives a lower limit for the cascade energy. Figure 8 gives some examples of very energetic cascades. We see that the corresponding cascade energy estimates lie above T_m , if we use $\xi = .3$. However, the values for the lower limit in cascade energy $E_{\min}$ ($\xi = 1$) still are rather high. We presume that some of these cascades might actually have had cascade energies above T_m . This is possible if for example an energetic heavy particle (i.e. α or heavier) is emitted from the compound nucleus.

ANALYSIS

We shall now compare our results with the number densities and energies of the cascades that one expects from 14 MeV neutron bombardment of Au. To this end we shall calculate the recoil energy distributions and cross sections for a 14 MeV neutron bombardment. Together with the known criteria for cascade cluster formation one can then make a direct comparison between the experiment and the theoretically predicted number densities and sizes.

Probability function for cluster formation

From charged particle irradiations we know the conditions under which visible clusters are formed. One finds that the probability to form visible cluster increases from 0 to 1 as the cascade energy goes from 0 to 50 keV. Above 50 keV always at least one cluster is formed. The probability to see at least one cluster per cascade is plotted in Fig. 9 as a function of cascade energy. Two curves are shown in Fig. 9. The solid curve was derived indirectly from charged particle irradiations in the Rutherford range,⁴⁾ the dashed curve is from values determined in self and heavy ion bombardments.⁷⁾ We shall see that this difference in the threshold functions is of minor importance for the purpose of the present calculations. In the following we shall base our calculations if not mentioned otherwise on the probability function obtained from the heavy ion results (dashed curve). It should perhaps be mentioned that some authors^{6,16)} have reported threshold curves that indicate a unit probability for cluster formation near 10 to 15 keV. These results have not been upheld and can easily be explained in terms of experimental difficulties encountered in heavy ion bombardments.⁷⁾

Elastic collisions

When a neutron is elastically scattered at a nucleus, the total kinetic energy is conserved and some of the kinetic energy of the neutron is transferred

to the nucleus. This recoil energy that the nucleus receives simply depends on the scattering angle θ in the center of mass system in the following way:

$$E_R = T_m \left(\frac{1 - \cos \theta}{2} \right) \quad (1)$$

where $T_m = \frac{4M_1M_2}{(M_1 + M_2)^2} E_n$ is the maximum energy that can be transferred in the collision, with M_1 , M_2 being the neutron and target mass and E_n the incident neutron energy. For 14 MeV n on Au we have $T_m = 281$ keV.

The differential elastic scattering cross section for 14 MeV neutrons on gold $\frac{d\sigma_{el}}{d\Omega}$ is shown in Fig. 10 as a function of $\cos \theta$ or, with the help of relation (1), directly as a function of the recoil energy E_R . The energy spectrum of the visible cascades is obtained by folding the probability function for cascade cluster production $W(E)$ with $\frac{d\sigma_{el}}{dE} = \frac{4\pi}{T_m} \frac{d\sigma_{el}}{d\Omega}$:

$$\frac{d\sigma_{el}^c}{dE} = \frac{4\pi}{T_m} W(E) \frac{d\sigma_{el}}{d\Omega} \quad (2)$$

$W(E) \frac{d\sigma_{el}}{d\Omega}$ is indicated in Fig. 10 by the dashed line and coincides with $\frac{d\sigma_{el}}{d\Omega}$ above 50 keV. The total cross section for the production of observable cascades is now given by

$$\sigma_{el}^c = \frac{4\pi}{T_m} \int_0^{T_m} W(E) \frac{d\sigma_{el}}{d\Omega} dE \quad (3)$$

Using for $W(E)$ the dashed and solid lines in Fig. 9 we find $\sigma_{el}^c = 4.3 \times 10^{-25} \text{ cm}^2$ and $5.6 \times 10^{-25} \text{ cm}^2$ respectively in conjunction with the differential elastic cross section of Fig. 10 as obtained from the ENDF/B files.¹⁷⁾ These data are mainly based on optical model calculations. As we did not have any experimental measurements of the differential elastic cross section on Au available, we also calculated the total elastic cascade cross section on the basis of some measurements

on Hg^{180} together with a rather arbitrary extrapolation to large scattering angles. This gave $\sigma_{el}^c = 6.7 \times 10^{-15} \text{ cm}^2$ in conjunction with the lower threshold curve (dashed line in Fig. 9). It seems that the uncertainty in the spectra causes a deviation of similar magnitude as the uncertainty in the threshold function. We see that in any case the calculated cross sections are considerably smaller than our experimentally observed ones. We conclude that the elastic neutron collisions can account for less than 20% of the cascades that are observed. The average cascade energy leading to a visible cluster in an elastic neutron collision is given by

$$\bar{E} = \frac{\int_0^T W(E) \frac{d\sigma_{el}}{d\Omega} E dE}{\int_0^T W(E) \frac{d\sigma_{el}}{d\Omega} dE} \quad (4)$$

From this we calculate $\bar{E} = 70 \text{ keV}$. This is the average total cascade energy for visible cascades produced by elastic events. As seen in Fig. 10, the elastic scattering is predominantly taking place in the forward direction, leading to small recoil energies. Point defects and point defect clusters can, of course, also be created in such events. We have disregarded those only because they are below the visibility limit of TEM. We can, however, estimate how much these events can contribute to the total damage. It turns out that only 20% of the total recoil energy available from elastic collisions goes into the formation of submicroscopic defects. This means that in regard to the overall damage production (including nonelastic events) low energy recoils ($\leq 30 \text{ keV}$) contribute only about 5%.

Nonelastic collisions

Collisions in which the kinetic energy is not conserved are called non-elastic collisions. The nonelastic collisions include a great variety of different types of events. The most frequent ones are the ones, where one or two neutrons or a charged particle is emitted. Upon absorption of the incident neutron the nucleus receives a recoil energy $E_R = \frac{T}{4}$. Due to the extremely short lifetime of the compound nucleus we can assume that none of this energy is dissipated in displacements before the secondard particle(s) is(are) emitted. In this case the impulses from the two (or more) recoils are added and the resulting total recoil energy may be larger or smaller than $\frac{T}{4}$. The emission of the secondary particle is usually isotropic in the center of mass system.¹⁹⁾ The average recoil energy for the emission of one particle is then approximately given by

$$\bar{E}_R = \frac{1}{M_2} (E_n + M_3 E_3) \quad (5)$$

where M_2 is the target mass number, M_3 and E_3 the mass number and energy of the emitted particle, respectively. Because the emitted particle has usually a rather low energy (less than 3 or 4 MeV),²⁰⁾ it is very improbable that the two momenta cancel or give a low recoil energy. We can in fact show from our data that most of the inelastic recoils are energetic enough to produce a visible cascade. If we take our experimentally observed total cascade cross section σ_c and subtract from this the cross section due to elastic collisions, we get

$$\sigma_c - \sigma_c^{el} = (2.7 \pm .6) \times 10^{-24} \text{ cm}^2.$$

This is within experimental error, equal to the total nonelastic cross section $\sigma_{\text{nonel}} = 2.44 \times 10^{-24} \text{ cm}^2$. In first approximation we, therefore, can say that all of the nonelastic events produce a visible cascade. This seems quite reasonable, because the absorption of the primary neutron results in a recoil energy of $T_m/4 = 70 \text{ keV}$. The secondary particle would have to have in the average a momentum such as to reduce the total recoil energy to below 30 keV. These events obviously must have a very low probability. Nonelastic 14 MeV neutron events in Au are mainly due to the $(n,2n)$ reaction.²¹⁾ We have not made any detailed calculations of the recoil energy spectrum that might be expected from nonelastic collisions. Robinson²²⁾ and Logan²³⁾ have made some calculations on Nb on the basis of simple nuclear evaporation models. We might point out that the most probable energy for E_2 in these models lies near zero.²³⁾ Therefore, in nonelastic collisions the most probable recoil energy is $E_R = T_m/4 = E_n/M_2 \approx 70 \text{ keV}$. The average recoil energy is expected to lie somewhat above this value.

DISCUSSION

The present investigation has experimentally shown that very energetic displacement cascades are formed as expected from the large recoil energies that occur under 14 MeV neutron bombardment. In addition to this, the splitting into subcascades has become quite evident. The conclusion, that most of the damage is due to nonelastic n-scattering events is also of considerable importance, because of the fact that practically all of these events lead to very energetic cascades. We shall first discuss the implications of the rather large cascade energies.

Subcascade formation

The high cascade energy most obviously manifests itself in the splitting of the cascade into subcascades. This splitting into subcascades so far has only found relatively little attention in discussions of cascade theory.^{24,25)} However, we would like to emphasize that the subcascade formation is quite important for the amount and type of damage that is generated in energetic collision cascades. Subcascade formation is expected whenever the mean free path between large energy transfers of the primary atom (or a secondary one of similar energy) is larger than the diameter of the subcascades and if the defect density along the path between large energy transfers is small compared to the defect density in the subcascades. As the cascade energy is increased, this situation always arises at some point simply from the fact that the average defect density in a cascade decreases with increasing energy²⁶⁾ and the average damage production along the path also decreases with increasing energy. Although there are some indications that channeling plays a role in the separation between subcascades, this is by no means a necessary condition for appearance of subcascades. Compared to gold the splitting into subcascades in Nb for example is expected to take place at lower energies. The fact that in Cu the subcascade clusters are usually not visible by TEM has been explained in this way. 5)

The subcascade formation also seems to be responsible for the fact that the clustering efficiency does not vary with damage energy as is observed in our heavy ion work. For stable clusters to be formed at finite temperatures we have to have sufficient spatial separation between vacancies and interstitials in order to have a high survival probability of clustered defects. This separation takes place in the form of replacement sequences²⁷⁾ that have a finite length. If the cascade would not split into subcascades, one would expect a

strong reduction in the defect survival with increasing cascade energy, once the cascade has reached a size that is comparable to the average length of the replacement sequences. With the introduction of subcascades, the basic subunit of the cascade does not change. With increasing energy we only get more of them. In the space between subcascades the concentration of interstitials will be higher than at the periphery of a single subcascade. This might lead to the formation of interstitial clusters considerably larger than those expected to be formed near the edge of individual subcascades. So far we have no evidence for visible interstitial clusters. At present, it is doubtful whether they could be identified as such by TEM contrast analysis. The strain field of the surrounding vacancy clusters would in any case produce very strong perturbations which would make a contrast analysis extremely difficult.

As the cascade energy is increased, an increasing fraction of the total cascade energy is lost to the electronic system and is not available for producing displacement damage. In gold, the electronic losses only amount to about 1/3 of the total cascade energy at 280 keV, if we calculate the electronic losses according to Lindhard²⁸⁾ and the formula given by Robinson.²²⁾ In lighter substances the electronic losses in 14 MeV neutron cascades are expected to be considerably higher, due to the combined effect of higher recoil energies and higher electronic losses compared to cascades of the same energy as in gold. In the past, the simple Kinchin and Pease model has been used in many cases to estimate the damage produced in displacement cascades. According to this model all of the energy above $E - M_2$ (M_2 = mass number in keV) goes into electronic losses. It might be worthwhile pointing out that our heavy ion results as well as, partially the very energetic cascades depicted in Fig. 8 are a direct illustration of the fact that the cascade damage is increasing beyond this limit. For better estimates of the damage produced at high cascade energies the modified Kinchin and Pease model proposed by Robinson²²⁾ should be used.

Nonelastic collisions

Our results indicate that most of the 14 MeV neutron damage is due to nonelastic neutron scattering events. The average recoil energy is expected to lie somewhat above 70 keV. We have also seen that the average recoil energy for those elastic collisions that produce a visible cascade also lies at about 70 keV. In contrast to this we estimate, however, from the cluster size distributions that the average damage energy is 135 keV. The average damage energy can also be estimated from the average number of clusters per cascade. In heavy ion irradiations we found that the cluster yield increases rather linearly with cascade energy between 50 and 300 keV. We get the relation $\frac{N_s}{N_c} = \frac{E_R}{67\text{keV}} = \frac{E_D}{50\text{keV}}$. From this and the observed number of clusters per cascade in the 14 MeV neutron case we deduce an average cascade energy $E_R = 120$ keV and an average damage energy $E_D = 90$ keV. Although the estimate from the size distribution does not agree very well with the estimate from the cascade multiplicity, there seems to be a clear indication that the average cascade energy lies significantly above $\frac{T_m}{4}$. This is not readily understood at present. A detailed analysis of the recoil spectrum from nonelastic collisions including all of the reactions would, of course, be necessary to get a good theoretical value for the average recoil energy. Large average recoil energies could be obtained if emission of heavier particles (α etc.) plays a significant role or if correlations between the neutrons in the $(n, 2n)$ reactions result in recoil energies significantly above $T_m/4$.

The effect of nonelastic recoils on the damage production is not limited to 14 MeV neutrons. Some time ago we found from a calculation of recoil energy distributions for a fission reactor spectrum that about 1/2 of the visible cascades in Au were produced by inelastic (n, n') scattering events. ⁴⁾ In this case a large fraction of the total damage went into the production of submicroscopic defects. Those were mostly produced in elastic scattering events. The

importance of the nonelastic component regarding the overall displacement production was, therefore, not as great as indicated by the factor of 1/2. On the other hand, in the 14 MeV neutron irradiation we have a very small fraction ($\sim 5\%$) of the total damage produced in events that do not lead to the formation of a visible cascade. These events are mainly those due to the strong forward scattering in the elastic scattering. Mitchell et al.²⁹⁾ have also found differences between TEM observations on reactor neutron and 14 MeV neutron irradiated copper samples. In this case not only vacancy clusters from the depleted zone, but also interstitial clusters are seen and the differences observed are most likely due to a combination of spectral and dose effects.

The nonelastic neutron cross section at 14 MeV is roughly equal to the geometrical cross section of the nucleus for most medium heavy and heavy elements.²¹⁾ The elastic cross sections are usually of similar magnitude, however, the predominant forward scattering gives low energy transfers and results in a much lower damage effectiveness of the elastic collisions. As a result one ends up with almost all of the damage produced in very energetic cascades. These energetic cascades produce a high local defect density which is present to a large extent in the form of spontaneously formed cascade clusters (interstitial and vacancy clusters). At elevated temperatures the presence of these clusters can drastically change the defect reaction kinetics. For example, clusters must break up before mobile defect species are formed and cascade clusters can act as nuclei for larger agglomerates. Energetic displacement cascades can also have large effects on the re-resolution of gas bubbles and other small precipitates.

CONCLUSIONS

The present investigation has directly shown some of the important aspects of the primary damage under 14 MeV neutron bombardment:

1. Damage is mainly produced in the form of very energetic cascades.
2. Subcascade formation is very pronounced. Up to 5 subclusters per cascade have been observed.
3. The observed number densities of the cascades agree quantitatively with what is calculated based on heavy ion results and the elastic and non-elastic neutron scattering cross sections.
4. Recoils from elastic neutron scattering events produce only 20% of the observed cascades.
5. Nonelastic collisions dominate the damage production. Within experimental error every nonelastic neutron scattering event gives rise to a visible cascade.
6. The average cascade energy as estimated from the number of vacancies clustered and the average number of clusters per cascade, is noticeably higher than what is expected from a rough estimate of the recoil energies.

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FIGURE CAPTIONS

- Figure 1 Gold film irradiated with 50 keV Au^+ ions for a fluence of $\sim 5 \times 10^{10}$ ions/cm². Each ion produces a 50 keV cascade with a visible defect cluster. Kinematical near [001]. Magnification 280,000X.
- Figure 2 Subcascade formation as seen in 200 keV cascades produced by Au self ion bombardment. Fluence was 2.6×10^9 ions/cm². Magnification 280,000X.
- Figure 3 Irradiation geometry. Monitor foils were taped to the outside of the capsule at the positions indicated. Note that the Aluminum specimen holder is positioned slightly asymmetrical relative to the neutron source and monitor foils.
- Figure 4 Stereo electron micrograph of 14 MeV neutron damage in gold. Fluence $\phi = 7 \times 10^{15}$ n/cm². The neutron beam was incident at a shallow angle ($\sim 35^\circ$) through the bottom surface. Top and bottom surfaces are easily located from the intersections of the two small microtwins that go from top to bottom at an angle of 55° to the surface. Note the individual black spots at various positions in the foil as well as the defect bunches at the sites of very energetic cascades. Magnification 140,000X.
- Figure 5 Number density of defect clusters versus fluence.
- Figure 6 Size distribution of subcascade clusters under a) 14 MeV neutron bombardment and b) 250 keV Au^+ self ion bombardment.
- Figure 7 Number density of visible cascades versus fluence.
- Figure 8 Examples of very energetic cascades produced by 14 MeV neutron bombardment of Au. Magnification is 500,000X. As we go from left to right in a) and b) we estimate the number of vacancies to be $N_v = 5700, 1600, 2000, 1900, \text{ and } 2800$ for the 5 most prominent cascades. This corresponds to minimum cascade damage energies of $E_D = 400, 110, 140, 130 \text{ and } 200$ keV respectively. The cascade with $E_D = 400$ keV clearly lies above T_m , however, there is the possibility that we see here two closely overlapping cascades. c) this particular feature seems

FIGURE CAPTIONS (contd)

to indicate line up of clusters along a linear track. Stereo-TEM, however, reveals that those clusters do not lie on a straight line. Line up to cluster is observed very seldom. d) shows 3 different cascades of varying size.

Figure 9 Probability function for cascade cluster formation. The dashed curve is from monoenergetic cascades, while the solid curve is derived indirectly from the energy dependence in the Rutherford region. $W(E)$ indicates the probability that one or more defect clusters are formed in a cascade of energy E .

Figure 10 Differential elastic 14 MeV neutron cross section of gold obtained from the ENDF/B files. The dashed curve coincides with the solid one above 50 keV and indicates the recoil distribution of visible cascades produced by elastic scattering events.

Figure 1

Figure 2

Figure 3

Figure 4

Figure 5

Figure 6

Figure 7

Figure 8

Figure 9

Figure 10