CONF-841157--99

To be published in the Proceedings of the Materials Research Annual Meeting, Advanced Photon and Particle Techniques for the Characterization of Defects in Solids Symposium, held in Boston, Massachusetts, November 26-30, 1984.

CONF-841157--99

DE86 002272

THERMAL ANNEALING OF VACANCY AND INTERSTITIAL LOOPS
IN ION IRRADIATED COPPER

B. C. Larson, T. S. Noggle, and J. F. Barhorst

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

By acceptance of this article, the publisher or recipient acknowledges the U.S. Government's right to retain a nonexclusive, royalty-free license in and to any copyright covering the article.

MASTER

SOLID STATE DIVISION
OAK RIDGE NATIONAL LABORATORY
Operated by
MARTIN MARIETTA ENERGY SYSTEMS, INC.
under
Contract No. DE-AC05-84OR21400
for the
U.S. DEPARTMENT OF ENERGY
OAK RIDGE, TENNESSEE 37831

January 1985

THERMAL ANNEALING OF VACANCY AND INTERSTITIAL LOOPS
IN ION IRRADIATED COPPER*B. C. LARSON[†], T. S. NOGGLE[†], AND J. F. BARHORST^{**}[†]Solid State Division, Oak Ridge National Laboratory, Oak Ridge, TN 37831^{**}Emerson Electric Company, St. Charles, Missouri 63301

ABSTRACT

X-ray diffuse scattering has been used to study the thermal annealing of vacancy and interstitial loops in Ni-ion irradiated copper. The diffuse scattering formalism is reviewed and diffuse scattering measurements are reported on liquid-He temperature Ni-ion irradiated copper after annealing to 40, 275, and 300°C. Size distributions are presented for vacancy and interstitial loops after each anneal and the thermal-induced changes are discussed in terms of loop dissolution and coalescence.

INTRODUCTION

At ambient temperatures, defect clusters are the primary form of structural damage resulting from energetic particle irradiation of metals. Depending on the nature of the irradiation particles, the initial defect configurations can vary from randomly distributed vacancy-interstitial pairs as produced by electron irradiations, to large three-dimensional defect regions, densely populated with vacancies and interstitials, which are typically produced by fast neutron and heavy ion irradiations.

Our understanding is incomplete on the processes whereby cascades collapse and vacancies and interstitials aggregate to form clusters such as dislocation loops, stacking-fault tetrahedra, or voids [1,2]. Experimental investigations have proved to be difficult as a result of the small sizes of the clusters and the tedious nature of direct observation measurements such as transmission electron microscopy (TEM) [3]. Diffuse scattering methods have been developed which complement the direct observation methods and are capable of providing detailed information on clustered defects when scattering models are available [4,5,6]. In this study, we have made diffuse x-ray scattering measurements of the size distributions of vacancy and interstitial loops in Ni-ion irradiated copper and we have studied the thermal evolution of these loops during annealing to 300°C.

THEORY

The scattering cross-section for randomly distributed lattice defects is given in the single defect approximation by [4]

$$\frac{d\sigma(\vec{K})}{d\Omega} = (r_e F(\vec{K}))^2 |A(\vec{K})|^2 \quad (1)$$

where $\vec{K} = \vec{h} + \vec{q}$ is the scattering vector specifying the measuring point in reciprocal space in terms of a reciprocal lattice vector $\vec{h}$ and a vector $\vec{q}$ relative to $\vec{h}$ within the Brillouin zone. The product of the classical electron radius r_e and the scattering factor $F(\vec{K})$ represents the scattering

*Research sponsored by the Division of Materials Sciences, U.S. Department of Energy under contract DE-AC05-84OR21400 with Martin Marietta Energy Systems, Inc.

length per unit cell of the host material, and $A(\vec{k})$ is the scattering amplitude associated with the defect cluster. For the case of dislocation loops [5,6],

$$A(\vec{k}) = \sum_i e^{i\vec{k} \cdot \vec{r}_i} + i\vec{k} \cdot \vec{S}(\vec{q}) + \sum_j e^{i\vec{q} \cdot \vec{r}_j} (e^{i\vec{k} \cdot \vec{S}_j} - 1 - i\vec{k} \cdot \vec{S}_j) \quad (2)$$

The first term in Eq. (2) is the direct scattering from the atoms at $\vec{r}_i$ comprising the dislocation loop and the second sum over the lattice represents the diffuse scattering caused by the displacements $\vec{S}_j$ of the surrounding atoms from the regular lattice position $\vec{r}_j$. $\vec{k} \cdot \vec{S}(\vec{q})$ is the so-called Huang scattering term, where $\vec{S}(\vec{q})$ is the Fourier transformed displacement field. $\vec{k} \cdot \vec{S}(\vec{q})$ can be evaluated in closed form using the Fourier transformed Green's function and the double force tensor, and the second summation converges rapidly in r since only higher order terms in $\vec{S}_j$ are retained.

At small q ($q \ll 1/R$), $\vec{k} \cdot \vec{S}(\vec{q})$ dominates the scattering so that [4,5,7]

$$|A(\vec{k})|^2 \propto \frac{b^2 k^2 R}{q} \quad (3)$$

where b is the Burgers vector of the loop and R is the loop radius. At larger q ($q > 1/R$) the scattering intensity falls off roughly as [7,8]

$$|A(\vec{k})|^2 \propto \frac{b k R^2}{q^4} \quad (3)$$

and is referred to as "asymptotic" or Stokes-Wilson" scattering. This scattering arises from local Bragg-like scattering from the strongly distorted region in the immediate vicinity of the loops. Therefore, the compressed region above and below an interstitial loop gives rise to diffuse scattering at positive q and vacancy loops give rise to scattering at negative q as a result of lattice relaxation into the volume of the missing plane. The average falloff of the intensity tends to be $\sim 1/q^4$, so the scattering from these regions modulates the falloff rather than actually causing a peak in the scattering. However, a peak does appear at $q \approx \pi/4R$ in a plot of intensity $\times q^4$ versus q .

These qualitative arguments can be seen quantitatively in Fig. 1, which shows numerical calculations at the (222) reflection for faulted $\langle 111 \rangle$ dislocation loops on $\{111\}$ planes in copper. The solid lines depict scattering for 1, 2, and 4 nm interstitial loops and the dashed lines represent the scattering from vacancy loops of these sizes.

Fig. 1. Diffuse scattering calculations, in the "Huang" and "asymptotic" scattering regions for vacancy (dashed) and interstitial (solid) loops, scaled by q^4/R^2 .

The important aspects of these results are the well-defined peaks in the intensities that appear at q values inversely proportional to R , and the fact that the vacancy loops scatter on the negative q side of the reciprocal lattice point and interstitial loops scatter on the positive q side. These properties make it possible to determine concentrations and size distributions for vacancy and interstitial loops separately by fitting measured diffuse scattering data with such calculations treating the concentrations as a function of R as fitting parameters.

The experimentally measured intensity in symmetric scattering geometry for a nonhomogeneous defect distribution is given by

$$I(\vec{K}) = \frac{I_0}{\sin \theta} \sum_i c_i^0 \frac{d\sigma_i(\vec{K})}{d\Omega} \Delta\Omega \int_0^t \frac{c_i(t)}{c_i^0} e^{-2\mu_0 t / \sin \theta} dt, \quad (5)$$

where I_0 is the incident beam power, μ_0 is the linear absorption coefficient, $c_i(t)$ is the concentration of loops of radius R_i , and $\Delta\Omega$ is the solid angle subtended by the detector. t is the depth in the crystal, c_i is the loop concentration at some reference point of particular interest (say at the surface). If the size distribution as a function of depth can be assumed constant and the relative shape of the concentration profile is known (i.e. from the damage energy profile), $c_i(t)$ can be determined using this expression.

EXPERIMENT AND RESULTS

In this work, copper crystals were irradiated with a primary beam of 17.4 MeV Ni-ions that were selectively degraded in energy such that (in terms of damage energy) a uniformly damaged layer (110 eV/A) 2.4 μm thick was produced. The irradiations were carried out at 4 K and the specimens warmed to room temperature before measuring. The x-ray diffuse scattering measurements were made at 10 K (to reduce the thermal background scattering) using a 12 KW $\text{CuK}\alpha$ rotating anode generator and a linear position sensitive detector. Measurements were made along the [111] direction at the (222) Bragg reflection; data were collected on an unirradiated portion of the sample as well as the irradiated portion in order to make corrections for the Compton and thermal background scattering. Thirty-minute thermal anneals were carried out at 40, 275 and 300°C in a vacuum of 2×10^{-6} torr.

Figure 2 shows diffuse scattering measurements made after annealing of the irradiated copper at temperatures of 40, 275, and 300°C. In these measurements q was oriented along the [111] direction and the q 's are referenced to the (222) reciprocal lattice point. The data in Fig. 2 have been scaled by q^4 for correlation with the calculations shown in Fig. 1. Apparently both vacancy and interstitial loops are present in the sample as there exists nearly equal scattering intensity for positive and negative q . The shape of the scattering is somewhat different on the two sides as the maxima in the intensities are at differing q -values and tend to occur at lower q 's after the higher temperature anneals. The solid lines in Fig. 2 represent least-squares fitting of the measured data using Eq. (5) as weighted by q^4 . Scattering calculations for both vacancy and interstitial loops of radii 1.0, 1.5, 2.0, 3.0, and 4.0 nm were used in the fitting procedure. The resulting loop concentrations have been plotted in Figs. 3 and 4 where the loop concentrations are distributed over ± 0.5 nm so that the areas of the graphs provide the total loop concentrations.

Fig. 2. X-ray diffuse scattering from Ni-ion irradiated copper after anneals to 40, 275, and 300°C. The data are scaled by q^4 .

Separate size distributions are shown for the vacancy and interstitial loops in addition to the combined size distribution; note the change in scale for the combined distribution. At all three temperatures the vacancy loops have smaller average sizes, although both the vacancy and interstitial loop distributions coarsen in size during the annealing steps. Table I indicates the number of point defects in the loop distributions and the percentage of the point defects that were incorporated in each type of loop is shown as well. Since equal numbers of vacancies and interstitials are created by the irradiation, a ratio of 50/50 would be expected, assuming all of the point defects are clustered into loops.

Anneal	N_{PD}	$N_I(\%)/N_V(\%)$
40°C	$4.1 \times 10^{19} \text{ cm}^{-3}$	49/51
275	2.4	45/55
300	1.9	48/52

Table I. Total number of point defects, N_{PD} , and the relative percentage of vacancy and interstitials in the dislocation loop distributions.

DISCUSSION

The results presented here demonstrate the use of x-ray diffuse scattering in determining size distributions for vacancy and interstitial loops. The clear separation of the vacancy loop scattering from the interstitial loop scattering shown in Fig. 1 makes this unambiguous separation of the size distributions possible. The inverse- R dependence of the positions of the peaks in the intensities (as plotted in Fig. 1) leads to particularly good resolution of the small loops; note the large shift between the $R = 1 \text{ nm}$ and $R = 2 \text{ nm}$ peak positions. The calculations required to generate the scattering cross sections are not trivial, but the displacement fields can be calculated numerically using anisotropic elasticity [9] and the techniques for evaluating the scattering cross-sections have been discussed [4,5].

No comparison is made between size-distributions generated by TEM and the x-ray scattering results generated in this work. However, a direct comparison has been made for 60-MeV Ni-ion irradiated copper [6]. Those results showed good agreement in both the size distribution and the absolute

Start
typing
here

Fig. 3. Vacancy and interstitial loop size distribution for Ni-ion irradiated copper after 40°C anneal.

Fig. 4. Vacancy and interstitial loop size distributions for Ni-ion irradiated copper after 275 and 300°C anneals.

magnitude of the loop concentrations at the larger sizes, but the x-ray diffuse scattering method identified significantly more loops at the smaller sizes than were reported in the TEM observations. In as much as the smaller sized loops were identified to be predominantly vacancy type (as found here in Figs. 3 and 4, as well) it was concluded that TEM was not identifying all of the smaller vacancy loops in the presence of the other loops in the sample. This conclusion is supported by the fact that in a number of cases, TEM observations [10,11] have not identified as many vacancies as interstitials incorporated in the loop distributions. Since TEM can image loops down to 1 nm diameter [12], the implication is that in routine imaging of thick samples, the smallest loops in the interior of the material may not always be identified. The size distributions of Stathopoulos [13] for neutron and ion irradiated copper indeed show the vacancy loop size distribution peaking at ~1 nm radius, as suggested by the x-ray measurements both here and in reference [6].

The annealing results shown here are not complete enough to make a detailed quantitative analysis of the dislocation loop evolution; however, they indicate that vacancy loops anneal by preferential loss of the smaller loops and that the number of large interstitial loops tends to increase during annealing. These observations are qualitatively consistent with dislocation line-tension driven shrinkage of the vacancy loops (which has the largest effect on the smallest loops) and a combination of vacancy-interstitial recombination at interstitial loops together with interstitial loop coalescence as a result of glide and climb by the smaller loops. The coalescence of two small loops decreases the number of smaller loops while increasing the number of larger loops.

CONCLUSIONS

X-ray diffuse scattering has been demonstrated capable of providing detailed information on dislocation loop distributions. The usefulness of the x-ray technique for annealing studies of irradiation damage lies in its nondestructive nature and the inherently good statistical sampling of the defects. The x-ray and TEM techniques are complementary in nature and by cross-correlation of the results generated by the two methods additional information on defect distributions can be obtained.

REFERENCES

1. K. Kitajima in Point Defects and Defect Interactions in Metals, ed. by J. Takamura, M. Doyama, and M. Kiratani, University of Tokyo Press, Tokyo, 1982 p. 807.
2. M. T. Robinson, in Fundamental Aspects of Radiation Damage in Metals, ed. by M. T. Robinson and F. W. Young, Jr., (ERDA CONF-751006-PI, Oak Ridge, TN, 1976) p. 1.
3. B. L. Eyre and C. A. English in Ref. 1, p. 799.
4. B. C. Larson and W. Schmatz, Phys. Stat. Sol. (b) **99**, 267 (1980).
5. P. Ehrhart, H. Trinkaus, and B. C. Larson, Phys. Rev.
6. B. C. Larson and F. W. Young, Jr., in Ref. 1, p. 679.
7. H. Trinkaus, Phys. Stat. Sol. (b) **54**, 209 (1972).
8. H. Trinkaus, Z. Angw. Phys. **31**, 229 (1971).
9. S. M. Ohr, Phys. Stat. Sol. (b) **64**, 317 (1974).
10. J. Narayan and S. M. Ohr, J. Nucl. Mat. **85/86**, 515 (1979).
11. A. A. Pochettino and M. Iphorski, J. Nucl. Mat. **57**, 356 (1975).
12. Y. Shimomura, K. Kitagawa, Y. Takai, and H. Hashimoto in Ref. 1, p. 709.
13. A. Stathopoulos, Phil. Mag. **44**, 285 (1981).