SATURATION AND RECOVERY IN NEUTRON-IRRADIATED MOLYBDENUM*

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A high-temperature (~70-1200°C) resistivity recovery study of fast-neutron-irradiated ($E_n \geq 1$ MeV) molybdenum shows an anomalous saturation effect above about (5-10) $\times 10^{19}$ n/cm^2. This effect is interpreted as resulting from the self-annealing of the stage-III defects at the higher fluences (~10^{21} n/cm^2) leaving essentially only the stage-IV defects after reactor ambient irradiation.

A number of studies have been reported in the past several years on the recovery of damage induced in molybdenum by neutron irradiation.[1-10] However, these studies, which have involved a number of techniques, have, in general, either only covered a limited neutron-fluence range or a limited recovery-temperature range. Therefore, a study was initiated to investigate the high-temperature recovery of molybdenum after high-fluence neutron irradiations. The important information desired was a clear picture of the high-temperature recovery phenomena and well-defined temperatures for the observed recovery stages. The method chosen for the observation of the induced damage and recovery was the isochronal resistivity technique. Preliminary results are presented in this paper; a more complete description will appear elsewhere.[11]

Six specimens were irradiated at reactor ambient temperature (~70°C) to fast-neutron ($E_n \geq 1$ MeV) fluences ranging from 1.1×10^{19} to 1.5×10^{21} n/cm^2. For every fluence a duplicate specimen was run. Since no significant differences were found, these are not presented here. The dependence of the irradiation-induced resistivity increment $\Delta\rho$ on the fast-neutron fluence (for the present studies as well as those of Peacock and Johnson[1]) is presented in Fig. 1. The resistivity increment $\Delta\rho$ is defined by $\rho_I - \rho_0$, where ρ_I is the as-irradiated resistivity, and ρ_0 is the preirradiation resistivity. The resistivity increment is seen to reach a maximum at between 5×10^{19} and 1×10^{20} n/cm^2, and then it decreases by about a factor of two at 1.5×10^{21} n/cm^2. This result is somewhat unexpected in view of the asymptotic approach to saturation considered in most investigations.[12-14] The slope of the linear portion of the curve (a slope of about 0.58) is in good agreement with that reported by Peacock and Johnson[1] (0.54). However, they observed what appeared to be somewhat similar peaking of the resistivity at about $(3\text{-}4)\times 10^{18}$ n/cm^2; the results of our studies as well as those of Kissinger, Brimhall, and Mastel[2] yield pronounced saturation effects at much higher fluences than Peacock and Johnson. Kissinger, Brimhall, and Mastel observed a saturation behavior similar to ours slightly below our fluences; however, these differences can be related to the different techniques of observation (x-ray parameter and length changes) utilized by the previous investi-

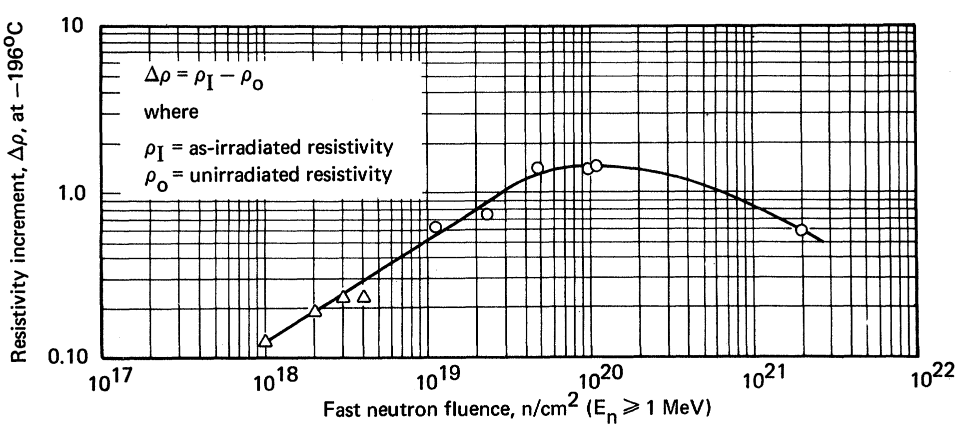

FIG. 1. Total radiation-induced resistivity increment for molybdenum as a function of fast-neutron fluence (dotted circles, data taken in this study; dotted triangles, Ref. 1).

gators.[11]

The recovery of the irradiation-induced defects in molybdenum is presented in Fig. 2 in which the radiation-induced resistivity increment $\Delta\rho$ is plotted versus the annealing temperature. The recovery of the irradiation-induced defects after reactor ambient (~70°C) irradiations resembles that for tungsten.[15] Two principal recovery regions are evident in all but the highest fluence specimen. Two principal recovery regions, centered at about 160 and 620°C, respectively, occur at almost exactly $0.15T_m$ (stage III) and $0.31T_m$ (stage IV) and agree quite well with the recovery observed by Ibragimov, Lyashenko, and Zavyalov[3] after a 2×10^{20}-n/cm^2 irradiation with fast neutrons. (T_m is the absolute melting temperature.) The lower temperature recovery region ($0.15T_m$) is believed to represent the migration and recovery of self-interstitials.[10,15] The term interstitial will be considered to mean self-interstitial unless specified otherwise.

Recent field-ion microscope studies by Jeannotte and Galligan[16] have shown that single vacancies migrate at about $0.31T_m$. It is therefore felt that vacancies also migrate in the stage-IV recovery region in molybdenum. Recent theoretical calculations have indicated that divacancies and some trivacancies in bcc metals (iron) also may have a migrational energy similar to monovacancies[17]; however, they would be expected to have quite different frequency factors for migration. Other smaller recovery regions appear between stages III and IV in some of these specimens, similar to tungsten[15]; however, the reproducibility of these smaller peaks is somewhat uncertain in our studies and the studies of others.[1,3,4] Their possible importance should not be overlooked since the defects responsible for these peaks apparently do contribute to the thermal hardening in this recovery region.[18]

The high-fluence specimen shows only a stage-IV recovery, indicating no stage-III recovery. The lack of a stage-III recovery can be interpreted as indicating a complete irradiation annealing of the interstitials (due to annihilation at vacancies and other interstitial traps as well as the formation of a large number of interstitial clusters). These results are in relatively good agreement with those of Kissinger, Brimhall, and Mastel[2] who have investigated by lattice-parameter and length-change studies the fast-neutron irradiation-induced damage fluence dependence on molybdenum over a range from 10^{19} to 10^{20} n/cm^2. The results of the length-change study indicate that vacancies saturate at about 10^{20} n/cm^2. The formation of an increased number of interstitial clusters (i.e., increase in the ratio of the number of interstitials in clusters to the number of free interstitials) up to a fast fluence of ~10^{21} n/cm^2 can satisfactorily explain the relative decrease in the stage-III recovery with increasing fluence. This increase in the relative

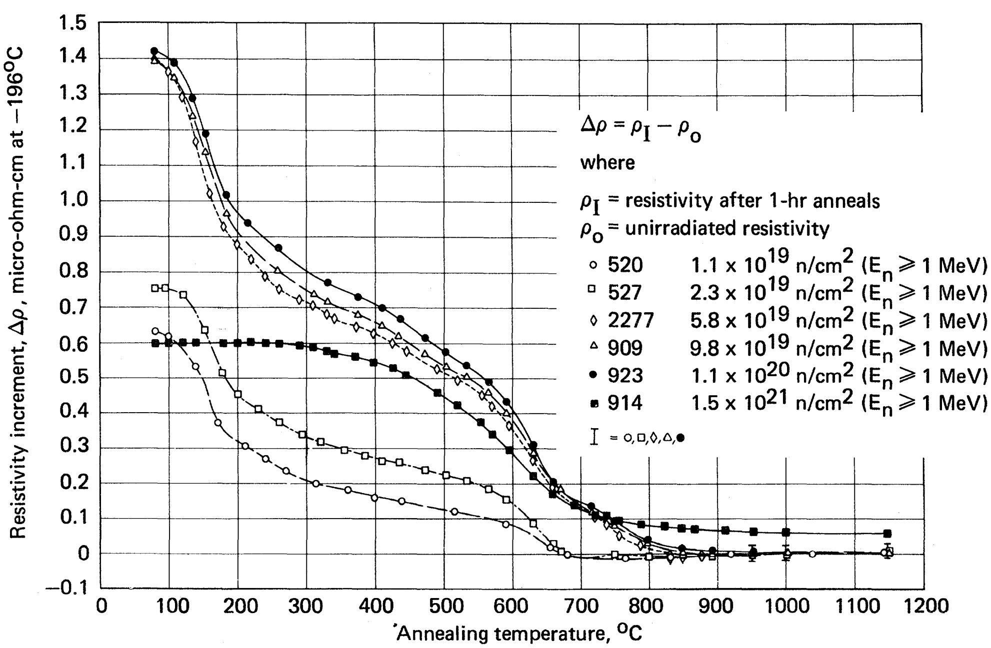

FIG. 2. Isochronal resistivity recovery of neutron-irradiated, recrystallized molybdenum as a function of annealing temperature.

number of large clusters with increasing neutron fluence is essentially the explanation given by Kissinger, Brimhall, and Mastel.[2] There are practically no free interstitials remaining at high levels of neutron fluence.

While it is conceivable that a higher irradiation temperature (200-300°C) could lead to the observed lack of a stage-III recovery for the highest fluence specimen, two principal factors eliminate this possibility. For a number of tungsten specimens, irradiated with the molybdenum specimens described here, no temperature effects were evident in the recovery curves.[15] That is to say, no flattening of the recovery above 70°C was observed. A more detailed analysis of the molybdenum recovery spectra[11] also indicates that stage III makes a definite contribution to the recovery with increasing fluence. A plot of the ratio of the recovery of stage III to the recovery of stage IV indicates a projected ~zero (or negligible) stage-III contribution in the range of 10^{21} n/cm^2 ($E_n \geq 1$ MeV). It is therefore felt that an irradiation temperature higher than 80°C for the highest fluence specimen can be ruled out.

The large differences in the recovery behavior observed between tungsten and molybdenum undoubtedly arises from the differences in the homologous temperature of irradiation.[15] For tungsten, the irradiation temperature of ~70°C corresponds to about $0.09T_m$. For molybdenum, however, the irradiation temperature of 70°C is about $0.12T_m$. This represents an irradiation within the stage-III recovery region in molybdenum and thus considerable recovery can occur during the irradiation. The recovery actually appears to begin at around 0°C.[4,19] The $0.09T_m$ irradiation temperature for tungsten is well below the recovery region for stage III[4]; thus from the recovery point of view, a larger number of point defects would be expected. Apparently irradiation within a recovery stage generates a larger ratio of cluster defects to point defects for the defects recovering in that recovery stage. This explanation is supported by both the much lower radiation-induced resistivity increment values observed for molybdenum than for tungsten at the same irradiation fluence, and the smaller slope observed for the dependence of these resistivity increment values on the neutron fluences in molybdenum.[15] These results are also in qualitative agreement with recent theoretical calculations of Beeler[19-21] concerning the influence of high fluence, high irradiation temperature, and displacement spike overlap on the damage state (in iron) after neutron irradiation. In particular,[19] these calculations show that at high irradiation temperatures (i.e., temperatures at which interstitials are mobile) the clustering of interstitials occurs quite rapidly due to rapid annealing in the displacement spike and that this leads to the existence of interstitial clusters even though clusters of more than two interstitials are rarely directly produced by a collision cascade.

In Fig. 2 it can be seen that a residual resistivity remains after the high temperature (1500°C) anneal of the specimen irradiated to the highest fluence. This residual resistivity is thought to result from the presence of technetium[22] produced via thermal neutron (n,γ) reactions.

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COHERENT EXCITATION OF POLARITONS IN GALLIUM PHOSPHIDE*

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The coherent excitation of polaritons has been achieved in the III-V semiconductor GaP for the first time by a nonlinear, optical parametric process. We are able to describe the polariton dispersion curve in the ω-k space and to measure the absorption coefficient of the crystal near the Reststrahl band. The results are discussed on the basis of recent theories on the polariton disperson and damping in GaP.

A high-intensity, coherent polariton field has been excited in the polar III-V semiconductor gallium phosphide by mixing two coherent optical waves whose wavelengths lie in the transmission gap of the crystal. This method for the creation in a medium of coherent, Raman-active optical phonons or other elementary excitation is sometimes called "coherent excitation[1]."

Four pairs of interacting frequencies have been chosen in order to excite resonantly the polarization field near the lattice resonance (Reststrahl) at 366 cm^{-1} covering, in the lower branch of the optical dispersion curve, a range of infrared wavelengths in which the excitation exhibits a mixed (electromagnetic and phonon) character.[2,3] In our experiment (Fig. 1) two Raman cavities pumped by a Q-switched ruby laser were used to generate two (Stokes) beams of coherent light at different frequencies (ω_1 and ω_2). The two beams were focused by a common high-quality achromatic lens (20-cm $f/1$) in the crystal where the polariton field at frequency $\omega_q = \omega_1 - \omega_2$ is generated. Small diaphragms were used in front of the lens in order to reduce the convergence of the beams reaching the crystal to about 5×10^{-3}. The angle θ made by the two beams $\vec{k}_1$ and $\vec{k}_2$ in

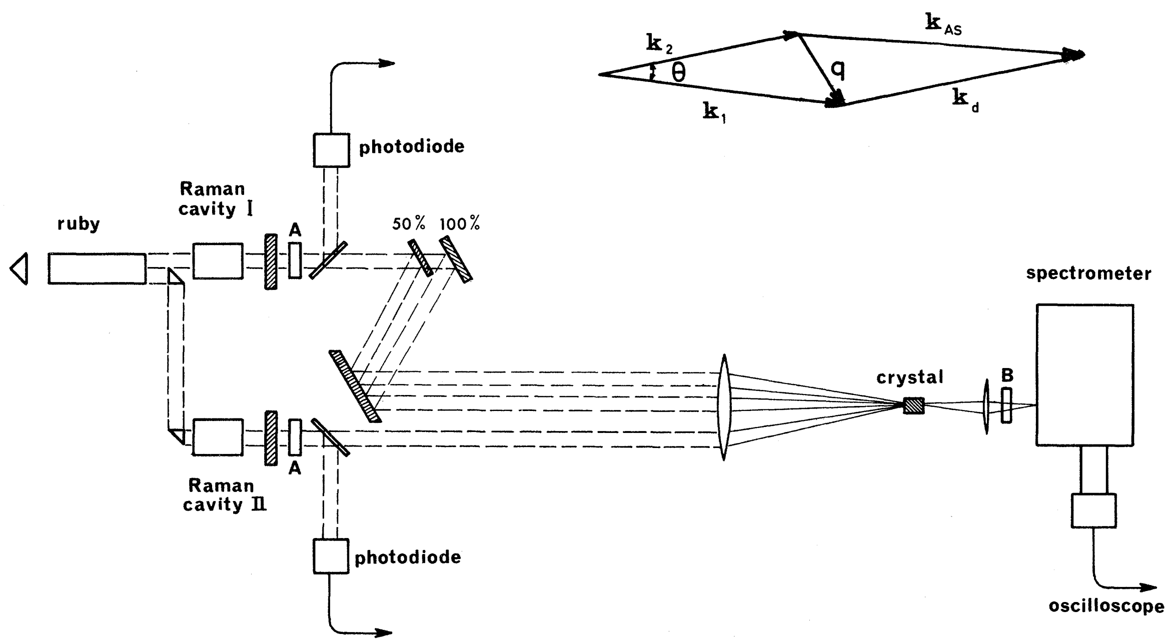

FIG. 1. Schematic diagram of the experimental apparatus.