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ELASTIC AFTER EFFECT STUDIES OF MOLYBDENUM AFTER ELECTRON IRRADIATION AT 4.7 K

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Abstract.- The self-interstitial in bcc Mo is expected to have the \(<110>\) dumbbell configuration. These dumbbells should therefore be able to reorient and eventually migrate simultaneously similar to \(<100>\) dumbbells in the fcc-metals. Because of the large shape parameters \(\delta\) of the self-interstitial in Mo as determined from Huang-scattering experiments, they should give rise to a large anelastic relaxation process. The identification of this process was the main aim of the present work.

A relaxation process with the expected properties has however not been found after low temperature electron irradiation. It is concluded that the self-interstitials in Mo undergo a two dimensional migration without reorientation.

Introduction.- The mechanical relaxation spectrum of Mo irradiated with fast neutrons shows two dominant peaks at 8K and 26K at 2 min relaxation time, which were attributed to the rotation without migration of the \(<110>\) dumbbell self-interstitial and of the di-interstitial, respectively /1/. No relaxation process indicating the simultaneous migration and reorientation of a dumbbell like that of the \(<100>\) dumbbell in electron irradiated Al /2/ could be detected. The shape parameter \((\lambda_1-\lambda_2)\) and \(\left(\frac{1}{2} (\lambda_1+\lambda_2)-\lambda_3\right)\) deduced for the 8K peak and the 26K peak are about one order of magnitude smaller than those determined for the self-interstitial atoms in electron irradiated Mo from Huang-scattering experiments.

The present elastic after effect studies on electron irradiated Mo were initiated in order to find out whether differences in the damage pattern after fast neutron and electron irradiation could account for these discrepancies and whether in electron irradiated Mo a process due to the simultaneous migration and reorientation jump of the \(<100>\) dumbbell would occur.

Experiment.- The elastic after effect measurements (EAE) were carried out on single crystalline cylindrical specimen in a torsional pendulum. Two differently oriented sorts of specimen were used, with the cubic \(<111>\) axis and the \(<110>\) axis along the torsion axis of the sample, respectively. The angular displacements were measured with a laser interferometer system with a resolution of \(\delta\phi = 10^{-5} \) rad. The electron irradiations were performed at the low temperature irradiation facility of KFA Jülich. The radiation induced defect concentrations were monitored by simultaneously...
irradiated electrical resistivity samples.

Results.- The results are shown in condensed form in fig. 1. The upper part (1a) shows the schematic relaxation spectrum, where the relaxation strengths is plotted versus the measuring temperature for a $<110>$-oriented sample. Four different relaxation processes have been identified in a narrow temperature regime between 22K and 28K after the electron irradiation at 300 ppm defect concentration. They are indicated by the $h$ bars in fig. 1a, whose position is at temperatures where the relaxation time $\tau = 100s$ and whose heights are given by the observed relaxation strengths. The orientation dependence indicates a $<110>$ orthorhombic defect structure for processes 2 and 3, and a $<100>$-tetragonal structure for processes 1 and 4. The activation energies and preexponential factors are in a range expected for jump processes of self-interstitials.

The next figure, fig. 1b, shows the annealing of the electrical resistivity during the isochronal tempering treatment. The differential curve, which shows the annealing stages more clearly, has been taken from ref. /4/ because of its better precision than the present one.

Finally, the four lowest curves fig. 1c-f show the behaviour of the $h$ peaks during the annealing treatment. In respect to the points under discussion given in the introduction the following items are important:

Fig. 1

Fig. 1a: Schematic view of the mechanical relaxation spectrum at $\tau = 100s$ for electron irradiated Mo. $\Delta_C$ is the maximum relaxation strength obtained during a 10 min isochronal annealing treatment. No processes below 20K and between 50K and 500K occur.

Fig. 1b: Recovery of the residual electrical resistivity (left scale) and differential recovery curve (right scale) taken from ref. /4/

Fig. 1c-f: Recovery of the different relaxation peaks 1-4 obtained in the isochronal Fig. 1f: annealing program.
1.) There is no relaxation process below 20K corresponding to the 8K-process detected in the neutron irradiated Mo /1/. There is also now additional relaxation process between 50K and 500K.

2.) None of the four peaks disappears in a unique annealing stage. Such a stage would indicate that the defect responsible for this process migrates or dissociates itself at that temperature. Apparently all 4 peaks are decreased partially by the defects responsible for the resistivity recovery peak at 28K, and their final decay is correlated with the recovery occurring between 35K and 42K.

3.) In order to compare the relaxation-strengths with those expected from the shape parameters observed in the Huang-scattering measurements, the sum of the relaxation strengths of all 4 processes is calculated and normalized to the initial Frenkel-pair concentration. The corresponding quantity can be calculated from the Huang-scattering results /3,5/. Both are given in table 1 for comparison, together with the corresponding data from the neutron irradiated Mo /1/.

<table>
<thead>
<tr>
<th></th>
<th>Present EAE</th>
<th>Huang /3/</th>
<th>n-irrad. EAE /1/</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\frac{1}{C_0} \sum_{i=1}^{4} \delta_i(c)$</td>
<td>17</td>
<td>1655</td>
<td>10</td>
</tr>
<tr>
<td>$\frac{1}{C_0} \sum_{i=1}^{4} \delta_i(c')$</td>
<td>38</td>
<td>1600</td>
<td>52</td>
</tr>
</tbody>
</table>

$\delta_i(c)$ and $\delta_i(c')$ are the partial relaxation strengths for $<100>$- and $<110>$-shear deformation, respectively. $C_0$ is the total defect concentration.

Discussion.- 1.) There is a clear difference between the EAE-spectra of electron and fast neutron irradiated Mo: The 8K peak is missing in the electron irradiated Mo, and instead of one unique peak at 26K a spectrum of four similarly large peaks is found around 26K. Because of the missing 8K peak the present measurements are not consistent with the interpretation of this peak in terms of a reorientation process of single $<110>$ dumbbells as proposed by Mizubayashi /1/.

Irrespective of question whether the reorientation of the $<110>$ dumbbell occurs with or without its simultaneous migration, if one of the processes could be attributed to the reorientation of the $<110>$ dumbbell, it should anneal in a stage like manner at the temperature where the dumbbell migrates. This has not been observed. The 8K and 26K-peak in the n-irradiated Mo as well as the present 4 peaks anneal all in a wide temperature regime and not in a unique stage.

2.) The shape parameters as determined by the Huang-scattering measurements are all-
most one order of magnitude larger than those determined from the present EAE-measurements, despite the fact that both measurements were made after electron irradiation of almost identical conditions. This can only mean that the dominant defects seen in the Huang scattering measurements are not or at most a minor fraction of them seen in the EAE-experiments.

Both points, the missing unique annealing stage of a relaxation process and the small relaxation strength as compared to the Huang scattering experiments lead to the conclusion, that the reorientation of the $<110>$-dumbbells in Mo is not observed by EAE-measurements.

Several arguments may be involved to account for this result: (i) The $<110>$-dumbbell needs less than 1 jump in order to disappear. In this case, also EAE-measurements would fail to observe this process. No such jump mechanism has however been proposed so far. (ii) A suppression of the expected relaxation process by mutual interaction of $<110>$-dumbbells may be excluded because this process is also missing in a sample irradiated to a Frenkel pair concentration of 10 ppm, at which concentration the interaction should be negligibly small. (iii) The $<110>$-dumbbell interstitial atoms may perform a migrational jump without reorientation. Among the jump processes proposed so far /6/ there is only the so called pure translational jump (fig. 2) which fulfills this requirement in principle. The activation energy $\tilde{H}^M$ for this jump is rather high. For the $\alpha$-Fe-potential of Johnson /7/ it is about 1 eV according to the calculations of Dederichs /8/. It appears rather unlikely that this jump could have such a low value as $\tilde{H}^M \approx 0.1$ eV in Mo as required to account for a migration occurring at about 30K to 40K /12,13/.

In spite of the lack of a suitable jump process we have reexamined the basic, characteristic resonant vibrations which the $<110>$-dumbbell may perform. As shown in fig. 3 there is one translational resonant mode, which gives rise to the translational jump, and there are two librational modes. A different kind of jump whereby a migration without reorientation of the dumbbell can occur may result from the stimulation of the librational I mode. This new jump model is shown in fig. 4. If the amplitude of the librational I mode is sufficiently large, one of the dumbbell atoms may jump over to the nearest neighbour atoms with which it forms a new dumbbell.
has the same orientation as the dumbbell before the jump. Clearly this jump leads to a two dimensional long range diffusion of the $<$110$>$-dumbbells in a $<$110$>$-plane without reorientation. From the calculations of Dederichs /8/ it is possible to give an estimate for the activation energy of this jump: since the saddle point configuration is very close the configuration of the $<$111$>$-dumbbell, one can take the difference of the formation energies of the $<$111$>$- and the $<$110$>$-dumbbells as a crude estimate. This difference is 0.28 eV for $\alpha$-Fe /8/. This value appears to be reasonably close to $H^M_0 \approx 0.1$ eV required by the experiments in spite of the ambiguities arising from the use of an $\alpha$-Fe potential in order to make an estimate for Mo.

If the $<$111$>$-dumbbell were the true (static) saddle point configuration, the present jump would only occur with a probability of 1/3 and jumps out of the particular $<$110$>$-plane with a probability of 2/3 due to the trigonal symmetry of the 111-axis. Because of the displacement pattern of the vibrational I mode, where also the next nearest neighbours are displaced strongly in the $<$110$>$-plane, the jumps in this plane may be highly favoured over the jumps out of the plane /9/. Such dynamical effects in diffusion phenomena are not unknown /10,11/. The present experimental observations require that the jumps within the $<$110$>$-plane occur with a probability of about 0.98. A theoretical estimate whether such a high degree of diffusion-anisotropy is realistic is not available at present. Future computer-simulation studies may be a possibility to give insight into such anisotropic diffusion phenomena.

The presently proposed new jump model explains also the unusual behaviour of resistivity annealing observed in Mo in stage I /12/ and of trapping of self-interstitial atoms at Fe$^{57}$-Mössbauer atoms /13/ on the basis of the two dimensional migration. Therefore these observations are another support of the present jump model.

**Conclusion.**—The comparison of EAE experiments on neutron irradiated Mo /1/ and of the present ones on electron irradiated Mo with Huang-scattering measurements on electron irradiated Mo /3/ leads to the conclusion, that the $<$110$>$-dumbbell self-interstitial atom in Mo does not reorient before or during its migration. This observation is explained by a model of a jump process which leads to a two dimensional migration of $<$110$>$-dumbbells in $<$110$>$-planes without reorientation.
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