DISLOCATION PINNING EFFECTS IN OXIDIZED AND REDUCED COPPER AFTER ELECTRON IRRADIATION AT 4.2 K

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Abstract - Oxidized and reduced copper samples were irradiated at 4.2 K with 3 MeV-electrons. The observed pinning stages in both states are compared and discussed. Also a comparison with former irradiation experiments is given.

I - INTRODUCTION

The difference in the pinning behaviour of oxidized and reduced copper samples has been investigated in the Hz-range after cold-work at about 100 K /1/ and after electron irradiation at 130 K /2/. In the present experiments such samples were irradiated with 3 MeV-electrons at 4.2 K and the internal friction was measured at about 2 kHz. In contrast to the Hz-experiments the annealing temperature of the samples had to be decreased to 1023 K /3/. The dislocation pinning due to the stages I_E, II and III can be seen for both states of the samples. In the oxidized sample annealed at 1173 K in stage II depinning is observed instead of pinning. The pinning and depinning effects are discussed and compared with other irradiation experiments.

II - EXPERIMENTAL PROCEDURE

A feed-back system was used to measure decrement and frequency at about 2 kHz /3/. The maximum strain amplitude was between 3×10^{-8} and 6×10^{-8} corresponding to the different amplitude dependences of both types of samples /3/. The polarizing voltage of 10 V caused a maximum static strain amplitude of 3×10^{-8}. The samples were annealed at 1023 and 1173 K /3/. The reduced samples (R) and the oxidized samples (O) were irradiated with 3 MeV-electrons at 4.2 K. A dose of about 5×10^{17} electrons/cm^2 was sufficient to reach the background values of internal friction.

III - EXPERIMENTAL RESULTS

For the reduced sample R.1023/1 (annealed at 1023 K), Fig.1 depicts the frequency (left ordinate and open symbols) and decrement (right ordinate

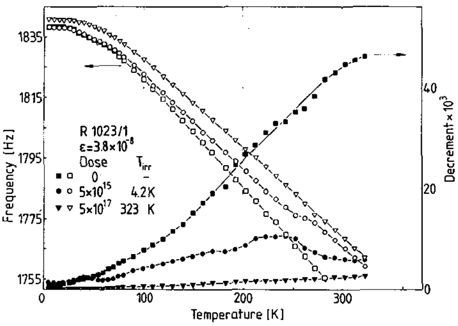

Fig.1: Temperature dependence of frequency (left ordinate and open symbols) and decrement (right ordinate and filled symbols) of sample R.1023/1 before irradiation (squares), after $5x10^{15}$ electrons/cm^2 (circles) and after $5x10^{17}$ electrons/cm^2 (triangles).

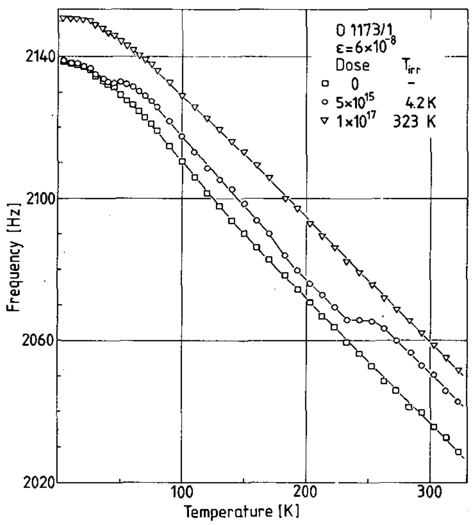

Fig.2: Temperature dependence of frequency for sample 0.1173/1 before irradiation (squares), after $5x10^{15}$ electrons/cm^2 (circles) and after $1x10^{17}$ electrons/cm^2 (triangles).

and filled symbols) as function of temperature. After 5×10^{15} electrons/cm^2 (circles) one can recognize at about 60 K the pinning stage I_E (more clearly in the decrement curve), between 110 and 220 K the pinning stage II and between 245 to 275 K the pinning stage III. At 300 K the modulus increase from the unirradiated (squares) to the background state (triangles, 5×10^{17} electrons/cm^2), e.g. the modulus defect, is 2.1 %. For the pinning stages the following mean pinning point numbers can be obtained: $p_I = 0{,}14$, $p_{II} = 0.16$ and $p_{III} = 0.84$ (see Table I). The temperature dependence of frequency for sample 0.1173/1 (annealed at 1173 K) is given in Fig.2 for the three states of the sample. Here exists a Bordoni peak /3/, as can be seen from the steeper slope between 50 and 150 K of the curves for the unirradiated (squares) and slightly irradiated (circles) sample in comparison to the background state (triangles). This steeper slope represents the modulus defect of the Bordoni peak. Therefore no p - value can be evaluated for

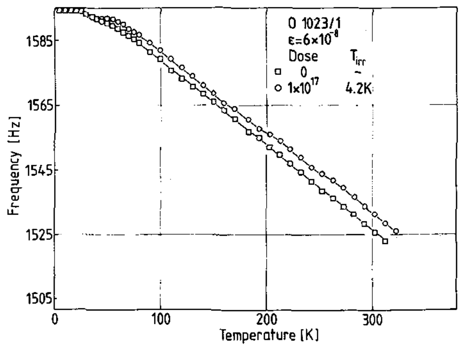

Fig.3: Temperature dependence of frequency for sample 0.1023/1 before irradiation (squares) and after background dose of 1×10^{17} electrons/cm² at 4.2 K (circles).

stage I_E, but this stage exists definitely. Instead of pinning stage II a modulus decrease between 170 and 190 K is found, e.g. depinning. But not all pinning that occurred in stage I_E recovers here. For stage III one obtains $p_{III} = 0.61$ and the modulus defect at 300 K is 1.2 %. The oxidized sample 0.1023/1 (annealed at 1023 K) was irradiated at 4.2 K with 1×10^{17} electrons/cm², corresponding to about the background dose (circles in Fig.3). As can be seen from Fig.3 the pinning stage I_E exists with $p_I = 0.22$, but from stage III only a small part can be seen because the background value of modulus is reached before the end of this stage. However, at this large dose no depinning is observed during stage II, but pinning stages appear at 165 and 193 K with $p_{II} = 0.48$. The modulus defect of this sample at 300 K is 0.9%.

III - DISCUSSION

(i) For the discussion of dislocation pinning it will be assumed that during irradiation at 4.2 K equal numbers of interstitials and vacancies are produced, and that in stage I_E freely migrating interstitials, in stage II interstitials released from impurity traps and in stage III vacancies pin the dislocations.

(ii) Even at the background dose at 4.2 K no pinning effects due to dynamic defect-producing processes ending at the dislocations could be detected (the error of frequency measurement was $\pm 5 \times 10^{-5}$).

(iii) Table I compares the pinning stages found in this work with those of other kHz-experiments /4,5/. Here for the irradiation at 78 K the spontaneous pinning during irradiation is ascribed to stage I_E /5/. One recognizes agreement in the temperature positions of the stages, only stage II for R.1023/1 is smeared out over a wider temperature range.

Table I: Comparison of different pinning experiments. E = electron energy, ϕ = electron dose, ϕ_∞ = background dose, T_{irr} = irradiation temperature T_i = temperature of pinning stage i and p_i = pinning point number in stage i.

	Keefer et al. /4/ (Fig.6)	Naundorf et al. /5/ (sample B in Fig. 2)	present authors (R.1023/1)
E	1 MeV	3 MeV	3 MeV
ϕ	4.5×10^{16}	4×10^{13}	5×10^{15}
T_{irr}	20 K	78 K	4.2 K
T_I	50 K	----	50 - 80 K
p_I	0.17	0.10	0.14
T_{II}	200 K	220 K	110 - 220 K
p_{II}	0.28	0.20	0.16
T_{III}	270 K	290 K	245 - 280 K
p_{III}	0.10	0.32	0.84
ϕ_∞	9×10^{16}	1×10^{17}	5×10^{17}

This will be certainly due to differences in the spectrum of trapping impurities.

(iv) For R.1023/1 the ratio $p_I/p_{II} = 0.9$ indicates that the fraction of interstitials arriving at the dislocations during stage I_E is about equal to that arriving in stage II after being released from impurity traps. This is in accordance with the high concentration of impurity atoms in the reduced sample /3/. The ratio of the pinning point number formed by interstitials to that formed by vacancies, that is $(p_I + p_{II})/p_{III} = 0.36$, is clearly smaller than one. Losses of interstitials may occur from the trapping at impurity atoms with high binding energy or from the simultaneous formation of interstitial clusters in the lattice, which can be more effective than the formation of vacancy clusters in stage III.

(v) While the p_I - values of Table I agree for all three experiments, there are larger deviations for p_{III}. In both other experiments the samples have been annealed under slightly oxidizing conditions. Thus this discrepancy can be attributed to the fact that always larger stages III are observed in reduced samples than in oxidized ones /2/. Also in the present work the ratio of p_{III} for 0.1173/1 and R.1023/1 is 0.62, which is in good agreement with results obtained for similar samples in the Hz-range /2/.

(vi) The pinning missing in stage II for 0.1173/1 corresponds to its higher residual resistivity ratio RRR = 1375, e.g. its smaller concentration of impurity atoms as possible trapping sites (for R.1023/1) one has RRR = 816). Thus it is possible to observe depinning between 170 and 190 K. This decrease of the number of pinning points formed in stage I_E was also found by Keefer et al. /4/ between 90 and 170 K. It agrees also with results obtained in the Hz-range after electron irradiation at 20 K /6/ (depinning between 130 and 190 K) and at 78 K /7/ (depinning between 120 and 180 K). In the last case there was a complete recovery of the pinning produced at 78 K. This difference to the present partial depinning, similar to that observed in /6/, can be explained by the assumption that only interstitial clusters can pin the dislocations and that there are different distributions of cluster size after irradiations below and above stage I_E /7/. After irradiation at 4.2 K the interstitials arrive at the dislocations in the narrow temperature range of stage I_E, i.e. their rate of arrival at the dislocations is higher than for the nearly 3 hours of irradiation at 78 K. This rate of arrival of interstitials should prefer the formation of larger clusters, while at 78 K only very small clusters, probably di-interstitials, are formed. The single interstitials resulting from the decay of the small clusters will then be lost at the nodes or impurity atoms acting as pinning points, and complete recovery of the 78 K-pinning appears. If also larger clusters exist after irradiation below stage I_E the depinning can be ascribed to the Ostwald ripening of these clusters /7/, where larger ones grow at the expense of the smaller ones, because the binding energy of a single interstitial is higher in the larger clusters. So the number of pinning points decreases by the decay of the small clusters and partial depinning is observed.

(vii) For 0.1023/1 no depinning is observed in stage II, but pinning stages at 165 and 193 K. The appearance of these stages is not due to the high dose in this case, but is typical for the lower annealing temperature of the oxidized samples. Stage II is found here also at lower doses. The ratio $p_I/p_{II} = 0.46$ for 0.1023/1 differs by a factor of two from the value 0.9 for R.1023/1 and indicates a larger contribution of stage II in the oxidized sample than in the reduced one. This is in contrast to the lower concentration of impurity atoms that can be expected from the higher RRR-value of oxidized samples. However, if only multiple-interstitials can act as pinning points, the higher dose for the oxidized sample will favour the formation of these clusters also after the release of interstitials from impurity traps in stage II.

(viii) Both 3 MeV-irradiations in Table 1 show about the same p_I-values and the total number of pinning points observed till 323 K differs only by a factor of about two, while the applied doses vary by two orders of magnitude. This discrepancy cannot be explained only by a difference in the mean free loop length of both experiments. There should also exist a loss of interstitials available for dislocation pinning if one irradiates below stage I_E. Then the high instantaneous concentration of mobile interstitials in stage I_E increases the probability of reactions between the interstitials, i.e. the formation of interstitial clusters in the lattice. Thus for a given dose the number of interstitials that arrive at the dislocations should be smaller than during an irradiation above stage I_E, where the instantaneous concentration of interstitials is much smaller and cluster formation in the lattice is not so probable. Besides this the higher rate of arrival of interstitials at the dislocations in stage I_E creates larger interstitial clusters as pinning points (see (vi)) than during an irradiation at 78 K. This also reduces the efficiency of pinning for a given dose, if it is applied below stage I_E. In accordance with the present work also for the irradiation at 20 K in Table 1 a higher dose was necessary. But here a factor of about 2 is due to the lower energy of the electrons /5/. However, the doses necessary to achieve the background values of internal friction are about the same (see Table I).

IV - SUMMARY

After irradiation of reduced and oxidized samples the well-known pinning stages are found. For stage II and stage III differences in size could be observed. In one case depinning was found in stage II that corresponds to other observations. However, the doses necessary at 4.2 K to obtain similar pinning effects are two orders of magnitude larger than in former irradiations at 78 K.

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