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DISLOCATION RELAXATION PEAKS IN TANTALUM, INTRINSIC AND IMPURITY DEPENDENT EFFECTS

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Abstract.- Investigations of the dislocation relaxation peaks in high purity and oxygen doped tantalum single crystals were carried out in the temperature range between 20 K and 700 K. The observed effects can be divided into 3 groups: the hydrogen Snoek-Köster relaxation (α^H) between 50 K - 250 K, the γ-relaxation between 350 K - 500 K, and an oxygen Snoek-Köster peak (S.K.P.) above 500 K for a measuring frequency of about 1 Hz. Only the effects above 300 K will be discussed here. The γ-relaxation consists of 2 subpeaks γ_1 and γ_2 which are related to the intrinsic dislocation motion and an interaction of dislocations with interstitial oxygen respectively. The S.K.P. is discussed with regard to a new suggestion of A. Seeger.

1. Introduction.- After plastic deformation of b.c.c. refractory metals several internal friction peaks are observed which are called α, β, and γ-peaks in the nomenclature of Chambers /1,2,3/. In the discussion of these relaxation effects it has always been a problem to identify the intrinsic dislocation effects. Intrinsic point defects and impurities can interact with the dislocations. In particular mobile impurities can interfere with the dislocation motion. In the case of the α-relaxation this situation is manifested by the long standing uncertainty concerning the role of hydrogen in the low temperature relaxation processes (see /4/ for further references). Looking for the kink pair formation in screw dislocations, i.e. the γ-relaxation, oxygen could play a similar role since for the case of tantalum even the highest purity samples contain small amounts of residual oxygen. For this reason there exists only little information about the γ-relaxation in tantalum. We report here on some results for the relaxation effects observed above 300 K.

2. Experimental procedure.- The experiments were carried out in an inverted torsion pendulum in the temperature range from 20 K to 700 K at a vibration amplitude of $\varepsilon=3\times10^{-6}$. The pendulum provided the possibility for "in situ" deformation of the samples at any temperature.

The tantalum single crystals were purified by annealing in ultra high vacuum at a pressure of $p<1\times10^{-7}$ Pa. The impurity concentration (O+N+C) was determined by measurement of the residual resistance ratio

RRR. After the above mentioned treatment the RRR was $\geq$5000. Some high purity samples were doped with 25 atppm oxygen as determined by the RRR and the height of the oxygen-Snoek peak. The hydrogen content of the samples was determined /5/ to be between 45 and 60 atppm.

3. Results for high purity crystals.- Figure 1 shows a typical spectrum as obtained after low temperature (T<375K) deformation. Below room temperature - from 50 K to 250 K - we see a group of effects which were formerly called the α-relaxation. By comparison with the results in niobium /4/, we consider this as a hydrogen-S.K.P. and call it α^H-relaxation in this paper. Using a similar method as Klam /6/, which was based on the assumption of symmetrical subpeaks on the 1/T-scale we can separate 4 subpeaks. These are called α_1^H through α_4^H beginning at the high temperature side of the α^H-relaxation. Out of these α_1^H and α_2^H are enhanced by high temperature deformation, α_3^H and α_4^H are enhanced by low temperature deformation. Above 300 K one notices first of all 2 stages of modulus defect recovery, which coincide with the onset of oxygen and nitrogen migration respectively. A second measurement run verifies that dislocations have been pinned to some extent by interstitial impurities. This, of course, hinders the observation of the γ-relaxation which falls into temperature range of oxygen migration.

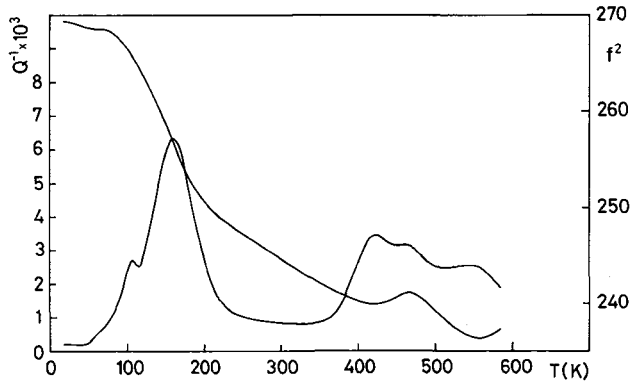

Fig. 1: High purity Ta single crystal [111], frequency 16 Hz, deformation:
3.1% tension at 375 K
+ 0.5% tension at 200 K
+ 0.2% torsion at 295 K

To see the aging effects more clearly we have conducted a series of measurements shown in figure 2. In this series the highest temperature reached during each run was increased by 25 K, starting at 400 K. The spectrum above room temperature contains 3 relaxation processes: the γ-relaxation, which consists of 2 subpeaks γ_1 and γ_2, and another process above 500 K, an oxygen S.K.P., as explained later. All maxima above 300 K have disappeared after annealing at 625 K except γ_2.

The overall concentration of O+N+C in our high purity specimens was less then 5 atppm. We could not specify, however, the oxygen concentration itself. To get a clearer picture of the role of oxygen, we

have done the same experiments with a sample containing a small, controlled amount of oxygen.

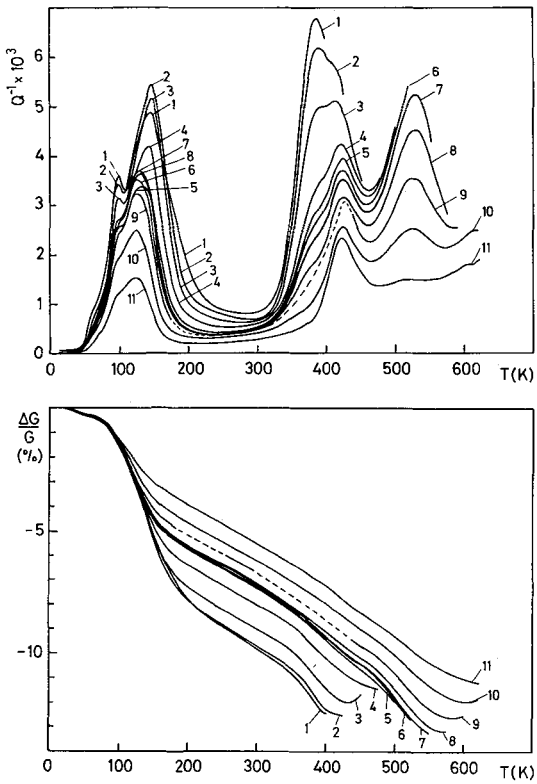

Fig. 2: Aging behaviour of damping and modulus after low temperature deformation, deformation history similar as in figure 1, high purity Ta single crystal [111], frequency 1 Hz, consecutive runs are shown

4. Results for oxygen doped crystals.- Figure 3 shows a spectrum of an oxygen doped sample in the same state of deformation as figure 1. The α^H-relaxation is smaller and shifted to lower temperatures as expected from the reduction of the free dislocation length due to the higher oxygen concentration. The more drastic changes, however, have occured

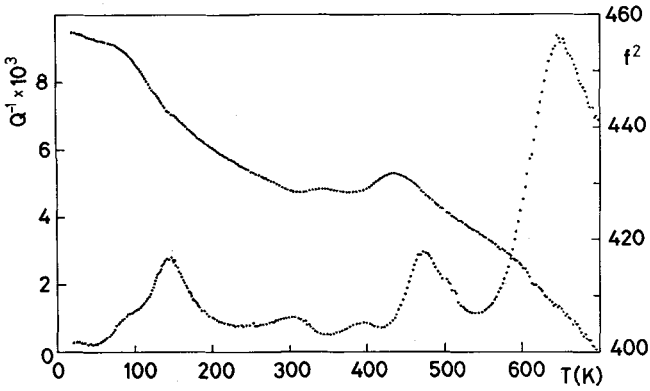

Fig. 3: Ta single crystal [111], 25 atppm oxygen, frequency 16 Hz, deformation:
3.6% tension at 372 K
+ 0.5% tension at 198 K
+ 0.2% torsion at 297 K

above room temperature. Starting at 300 K the modulus defect recovers which is manifested also by 2 "pseudopeaks" near 300 K and 400 K. At 475 K, about the same temperature as in the high purity sample, there is a large γ_2-peak. In addition there is a very large peak near 650 K.

The aging behaviour of the spectrum in a doped sample is shown in figure 4. The annealing out of the α^H-relaxation shows the arrival of oxygen atoms at the dislocations. A totally different behaviour has the

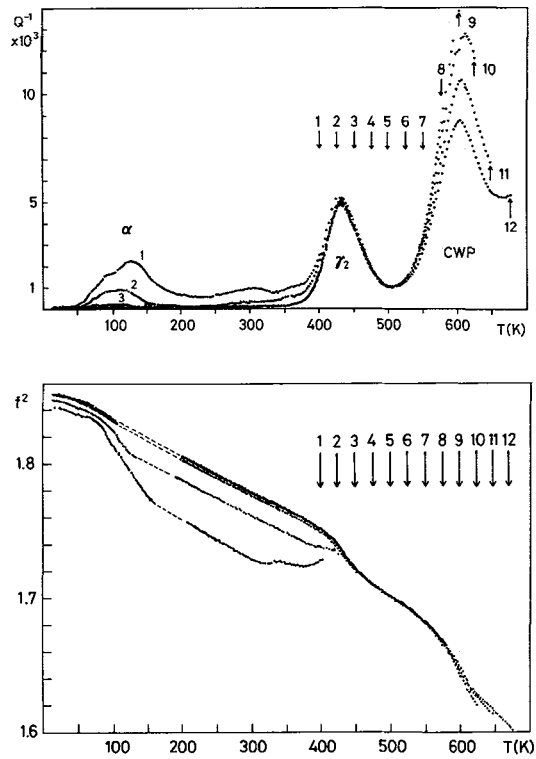

Fig. 4: Aging behaviour of damping and modulus after low temperature deformation, Ta single crystal [111], 25 atppm oxygen, frequency 1 Hz, all consecutive runs are shown

γ_2-peak, which has not dissappeared after anneals up to 650 K. The S.K.P. on the high temperature side of the γ_2-peak shows aging effects again. There exists no γ_1-peak in the doped specimen.

		γ_1	γ_2	S.K.P.
high purity tantalum	H (eV)	1.243 ±0.05	1.124 ±0.04	2.204 ±0.35
	$f_o(s^{-1})$	1×10^{16}	2×10^{13}	1×10^{21}
oxygen doped tantalum	H (eV)	--	1.170 ±0.04	2.242 ±0.17
	$f_o(s^{-1})$	--	6×10^{13}	5×10^{18}

Table 1: Activation parameters

Table 1 gives the activation parameters for the 3 processes γ_1, γ_2, and the S.K.P.. In the high purity samples the values were obtained after a separation of γ_1 and γ_2 by the same method as in the case of the α^H-relaxation.

5. Discussion.- The intrinsic dislocation relaxation is usually discussed in terms of 2 mechanisms: thermally activated kink diffusion and kink pair formation /2/. Because of the particular properties of screw dislocations in the b.c.c. structure /7/ the kink pair formation in screw dislocations is expected at temperatures T>room temperature for low frequency measurements; all other intrinsic dislocation effects take place below 300 K. Above 300 K one should therefor encounter basicly 1 intrinsic dislocation effect and dislocation - impurity interactions.

The height of the γ-relaxation increases after low temperature deformation. Because the relative density of screw dislocations increases after low temperature deformation /8/, screw dislocation segments seem to be involved in both subpeaks of the γ-relaxation. As to the dependence on deformation temperature there is also a correlation between the γ-relaxation and the low temperature components of the α^H-relaxation - α_3^H and α_4^H - which also seem to involve screw dislocation segments.

The 2 subpeaks γ_1 and γ_2 react much differently to the presence of oxygen atoms. When discussing this point one has to keep in mind that even in our high purity samples we might have an oxygen concentration up to 1 atppm. The arrival of oxygen atoms at the dislocations suppresses γ_1 (fig. 2) and higher oxygen concentrations can prevent the appearance of γ_1 from the start (fig. 4). In contrast to that is the increase of γ_2 with increasing oxygen concentration (cp. fig. 2 and fig. 4) and its stability with oxygen present at the dislocation line (fig. 4): the peakheight is stable or increases slightly as a function of annealing temperature in the doped as well as in the high purity samples. The activation parameters of γ_2 are very similar to those of the oxygen-Snoek peak (H = 1.1 eV, $f_o = 2 \times 10^{13}\ s^{-1}$ /9/). The γ_2-peak of figure 4 is 20 times higher than the oxygen-Snoek peak in the undeformed sample, i.e. plastic deformation enhances γ_2. It is impossible that this increase is due to the pick up of oxygen, because an increase in oxygen concentration by an order of magnitude should have shown up in the high purity samples very clearly.

The above observations lead us to the following interpretation: γ_1 is caused by kink pair formation in screw dislocations. γ_2 is caused by the motion of screw dislocation segments interacting with oxygen atoms. We therefor suggest the motion of oxygen decorated kinks in screw

dislocations as the underlying mechanism.

It is also interesting to see that in the doped sample the pinning stage due to oxygen atoms shows 2 substages: Snoek-ordering by long range interaction and diffusion of oxygen to the dislocation (Cottrell-pinning) /10/. This has also been observed by other authors /11/ and can be used as a method to compare the purity of degassed samples. The absence of the first substage in our high purity samples and the fact that pinning by oxygen atoms comes to an end before the complete suppression of the α^H-relaxation demonstrates the high purity of our samples.

For the maximum on the high temperature side of the γ-relaxation the predominant feature is its drastic shift to higher temperatures with increasing oxygen concentration; a behaviour as expected for a Snoek-Köster mechanism. This shift in peak temperature should come from the dependence of the preexponential factor on the impurity concentration at the dislocation line /12,13/. According to the theory of A. Seeger /13/ the activation energy should be independent of the impurity concentration and given by the following expression

$$H_{S.K.P.} \cong 2H_k + H_M \quad .$$

Here $2H_k$ denotes the kink pair formation energy of the involved dislocation and H_M the migration energy of the involved interstitial. Comparing the activation energies of γ_1, the oxygen-Snoek peak, and the peak on the high temperature side of the γ-relaxation this prediction is well fulfilled. Work on samples with dopings of different types of interstitial solutes (O,N,C) would be required to consolidate the validity of this prediction.

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