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HYDROGEN DIFFUSION IN TANTALUM

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Résumé. — L'effet Gorsky a été étudié dans le système H-Ta à très basse concentration d'hydrogène (0.12-0.25 at. %); les mesures ont été effectuées en utilisant des fréquences comprises entre 2,8 et 23 Hz. On a trouvé que le coefficient de diffusion de l'hydrogène est bien représenté par une loi d'Arrhenius entre l'intervalle de température 210-525 °K, les paramètres de diffusion étant $D_{0H} = 3 \times 10^{-4 \pm 0.6}$ cm²/s et $W_H = 0.15 \pm 0.03$ eV. Une déviation de la loi exponentielle a été observée à températures inférieures à 210 °K.

Abstract. — The Gorsky effect has been investigated in the H-Ta system at very low hydrogen contents (from 0.12 to 0.25 at. %); the measurements have been made in the frequency range 2.8-23 Hz. The diffusion coefficient of hydrogen has been found to obey an Arrhenius-type law in the temperature range 210-525 °K, the diffusion parameters being $D_{0H} = 3 \times 10^{-4 \pm 0.6}$ cm²/s and $W_H = 0.15 \pm 0.03$ eV. A deviation from the exponential behaviour has been observed at lower temperatures.

1. Introduction. — In recent years there has been great interest in the diffusion of hydrogen and deuterium in metals; this has taken place because experimental tools, such as inelastic neutron scattering [1-3], nuclear magnetic resonance [4-7], internal friction [8-14] and elastic after-effect [15-17], have recently been introduced in this field. The growing quantity of experimental results, showing the inability of the classical jump-rate theory to explain the isotope effect, has stimulated more and more refined quantum theoretical treatments [14, 18-27].

Although noticeable progress has been made in both experiment and theory, important questions are still open. One of these unsolved problems is the discrepancy found between the diffusion data in Nb [28], when obtained by the Gorsky effect or by a low temperature peak (LTP), which has been attributed by Cannelli et al. to a Snoek type mechanism. This discrepancy, in connection with some theoretical calculations [14, 19], has led some authors [27] to assume that the (LTP) may not be a Snoek-type relaxation process. A clarification of this point is of interest for the future development of the hydrogen and deuterium diffusion studies. This experimental work has been undertaken in an attempt to investigate the Gorsky effect of hydrogen in Ta and to compare the diffusion data so obtained with those deduced by Cannelli et al. from the (LTP).

2. Experimental procedure and results. — The internal friction measurements have been made in a series of thin sheets, prepared by cold-rolling and chemical polishing from 99.9 % pure Ta supplied by the Union Carbide Co. The final thicknesses *h* of the samples

Sample (No)	Thickness <i>h</i> (μ)	Inverse peak temperature $T_M^{-1} \times 10^3$ (°K ⁻¹)	Frequency at T_M^{-1} (Hz)
1	6	3.40	3.4
		3.38	3.4
		3.30	3.3
		3.30	3.6
		3.45	2.8
2	11	2.90	3.3
		2.85	3.4
		1.70	23

are listed in Table I, where are also collected the main data on the Gorsky relaxation effect. The samples have previously been recrystallized at about 1,250 °K for about 3 days, then electrolytically loaded with hydrogen and subsequently homogenized in vacuum at about 500 °K for three hours. Owing to the small dimensions of the samples, the content of hydrogen cannot be determined with good accuracy by the standard methods of analysis, so, indicative values have been deduced from the Gorsky relaxation strength in Ta (1.3×10^{-2} /1 at. % hydrogen at 300 °K [29]). The experimental technique used is that described in previous papers [11, 12].

The Gorsky effect is conveniently investigated by plotting TQ^{-1} as a function of T^{-1} . This quantity measured in specimen 1 after some subsequent hydrogen loading treatments, is plotted in figure 1, where is also reported the reference curve obtained after the recrystallization treatment. It can be seen that hydrogen impurities bring about a well-developed Gorsky

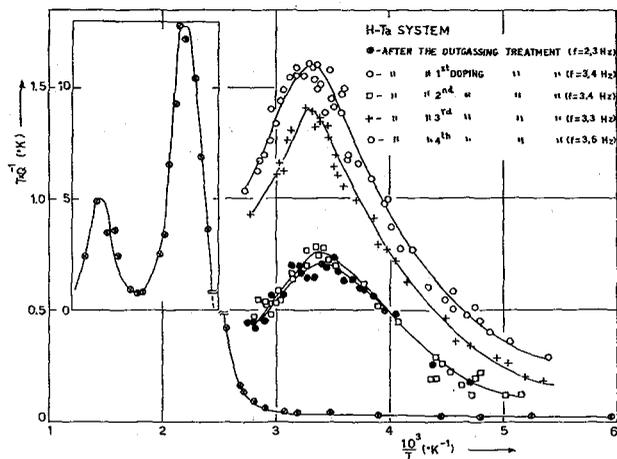


FIG. 1. — Internal friction in pure and hydrogen doped Ta.

peak which is followed, in the increasing temperature direction, by the oxygen and nitrogen Snoek effects. The maximum value of TQ^{-1} for the Gorsky peak is seen to increase with the loading time, the lowest value being 0.70°K . The hydrogen content corresponding to this value is about 0.12 at. %.

3. Discussion. — The internal friction due to Gorsky relaxation effect may be approximated by [30]

$$Q^{-1}(T) = \frac{\theta}{T - T_s} \frac{2\omega\tau}{1 + \omega^2\tau^2};$$

T_s is a characteristic temperature which depends upon the hydrogen content, $\tau = h^2/\pi^2 D$ the relaxation time, D the diffusion coefficient, ω the angular frequency of the vibration, θ a constant. For the above estimated content of hydrogen, T_s seems to be negligible over the whole temperature range of the measurements, so, no correction has been made on the data reported in figure 1.

The diffusion coefficient of hydrogen, as deduced both from the analysis of the relaxation curves (white points) and from the shift in the temperature scale of the Gorsky peak with frequency (black points), has been plotted in figure 2. Within the experimental errors, the two groups of points lie on the same straight line in the high temperature range, thus confirming that the Gorsky effect may be treated as a single

activated process. In the low temperature region the straight line does not interpolate the experimental data. The diffusion parameters evaluated from the straight line are

$$W_H = (0.15 \pm 0.03) \text{ eV};$$

$$D_{0H} = 3 \times 10^{-4 \pm 0.6} \text{ cm}^2/\text{s}.$$

In the insert of the same figure are also included values of $D(T)$, evaluated from data on the (LTP), assuming tetrahedral-tetrahedral jumps. It can be seen that in Ta, unlike in Nb, our high temperature data and those deduced from the (LTP) may be made to lie on the same straight line.

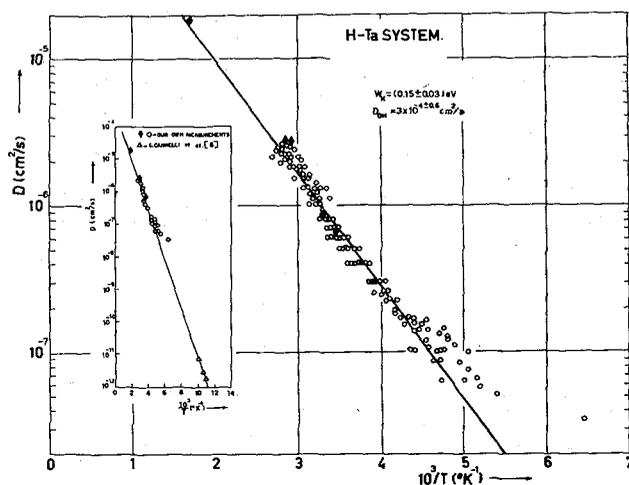


FIG. 2. — Diffusion coefficient of hydrogen in Ta; (\blacktriangle) from the shift of the peak with frequency; (\circ) from the analysis of the relaxation curves; (\triangle) from the low temperature peak (ref. [8]).

The interpretation of the experimental data in terms of a simple exponential law for $D(T)$ meets with the difficulty in explaining the deviation observed in the low temperature range of the measurements on the Gorsky effect. So, much more experimental work is needed both on the Gorsky and on the (LTP) relaxation effect before certain conclusions may be drawn.

Concluding, it can be noted that a less sure discrepancy exists between the Gorsky and the (LTP) diffusion data in Ta than in Nb.

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