Analysis of the Snoek relaxation in niobium–oxygen with low interstitial content

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Abstract

It is normally assumed in the literature that the Snoek relaxation in b.c.c. metals leads to internal friction peaks of the Debye type at very low interstitial contents. By means of a new measuring technique, however, it is shown that this is not generally the case. This technique allows simultaneous measurement of the internal friction *vs.* the temperature and of its derivatives with respect to the temperature and frequency. The data indicate that a distribution of relaxation times is present even for concentrations of a few hundred atomic parts per million. Some results for the Snoek relaxation in niobium are discussed in detail, showing that the parameters of the distribution change with the concentration of interstitials and the temperature.

1. Introduction

The dynamical response of the standard anelastic solid (SAS) leads to an expression for the internal friction F which is given by [1]

$$F = \frac{\delta}{(1+\delta)^{1/2}} \frac{1}{2 \cosh[\ln(\omega\tau)]} \tag{1}$$

where δ is the relaxation strength, τ is the relaxation time and $\omega = 2\pi f$, f being the frequency of the applied stress. It is assumed that τ changes with temperature according to the Arrhenius relation

$$\tau = \tau_0 \exp\left(\frac{H}{kT}\right) \tag{2}$$

where τ_0 is the pre-exponential factor, H is the activation enthalpy, k is Boltzmann's constant and T is the absolute temperature. Since δ depends only on the temperature, eqn. (1) can be written as

$$F = \frac{\alpha(T)}{2 \cosh\{\ln[\omega(T)\tau(T)]\}} \tag{3}$$

where $\alpha(T) = \delta/(1+\delta)^{1/2}$. An internal friction peak described by eqn. (3) is of the Debye type, *i.e.* a single relaxation process is involved. Two F and ω *vs.* T curves

measured in a torsion pendulum at a given moment of inertia do not give enough information to establish whether the internal friction peak can be described by eqn. (3). In fact, it has been shown [2] that even when a distribution of relaxation times is present, the internal friction can be described by

$$F = \frac{\alpha(\omega, T)}{2 \cosh\{\ln[\omega(T)\tau(T)]\}} \tag{4}$$

In a single experiment performed at a constant moment of inertia $\omega = \omega(T)$, so $\alpha = \alpha(T, \omega) = \alpha(T)$. In a torsion pendulum operating in free decay, once the specimen is fixed, F and ω depend not only on T but also on the moment of inertia, I, since by changing this last quantity, at a fixed temperature F and ω can be changed at will, even when $\tau = \tau(T)$ and $\alpha = \alpha(T)$. Then, strictly, $F = F(T, I)$ and there is an important partial derivative that should be considered in conjunction with the eqn. (4) [3], namely

$$\left.\frac{\partial \ln F}{\partial I}\right|_{(1/T)} = \left.\frac{\partial \ln \alpha}{\partial I}\right|_{(1/T)} - \tanh[\ln(\omega\tau)] \left.\frac{\partial \ln(\omega\tau)}{\partial I}\right|_{(1/T)} \tag{5}$$

This equation indicates that if $\alpha = \alpha(T)$ and $\tau = \tau(T)$, then

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$$\ln \tau = \tanh^{-1}\left(-\frac{(\partial \ln F/\partial I)|_{(1/T)}}{(\partial \ln \omega/\partial I)|_{1/T)}}\right) - \ln \omega$$

$$= \tanh^{-1}\left[-\left(\frac{\partial \ln F}{\partial \ln \omega}\right)\bigg|_{(1/T)}\right] - \ln \omega \qquad (6)$$

Then, by making small changes in the moment of inertia of the pendulum at each temperature and measuring the changes produced in F and ω, it is possible to obtain τ as a function of $1/T$. Furthermore, by additional changes in the value of I, it is possible to verify whether α depends only on T or also on I, i.e. to verify whether a single relaxation process or a distribution of relaxation times is involved in the internal friction.

Some internal friction data on the Snoek relaxation in Nb–O alloys with low interstitial contents will be presented to show that the Snoek peak is not of the Debye type. In addition, it will be shown how the parameters of the distribution of relaxation times change with the concentration of interstitials and the temperature.

2. Experimental procedures

The pendulum used and the measuring procedure have been described elsewhere [3, 4]. Briefly, at each temperature F is measured at a fixed moment of inertia in free decay, leading to F_1 and ω_1; subsequently, the moment of inertia is changed slightly to give new values for F and ω, namely F_2 and ω_2. According to eqn. (6), this procedure gives at each temperature

$$\ln \tau = -\ln \omega - \tanh^{-1}\left(\frac{\ln(F_1/F_2)}{\ln(\omega_2/\omega_1)}\right) \qquad (7)$$

The measurements were performed under high vacuum on a stress-relieved niobium specimen in the form of a sheet 29.3 mm long, 3.3 mm wide and 0.35 mm thick. Some additional measurements were performed on a specimen 10 mm long by using a torsion pendulum, with variable moment of inertia, operating at intermediate frequencies (of the order of 100 Hz) [5]. The purity of the material used is similar to that reported by Weller et al. [6, 7]. The concentration of oxygen in solution, C_O, in the specimens estimated according to the relationship [8]

$$C_O = \frac{F_p}{4.81 \times 10^{-6}} \quad \text{(at.ppm)} \qquad (8)$$

was of the order of 550 and 770 at.ppm. F_p indicates the maximum of the internal friction peak.

3. Results

The curves of Fig. 1 show the internal friction vs. the reciprocal of the temperature measured at three different moments of inertia, I_1, I_2 and I_3, in the specimen with 550 at.ppm O and two moments of inertia, I_1 and I_2, in the specimen with 770 at.ppm O. The procedure described by eqn. (7) was used at I_1 and I_3 in the specimen with 550 at.ppm O and at I_1 in the specimen with 770 at.ppm O to obtain $\ln \tau$ against $1/T$. The results are illustrated in Fig. 2, showing that $\ln \tau$ changes linearly with $1/T$, and a least-squares fitting to the data leads to the straight line indicated in the same figure and to

$$\tau_0 = 1 \times 10^{-14} \text{ s}, \qquad H = 1.11 \text{ eV} \qquad (9)$$

Then, in principle, τ depends only on the temperature and varies according to eqn. (2). There is an additional condition, however, that must be obeyed if the peaks of Fig. 1 are of the Debye type: α must be independent
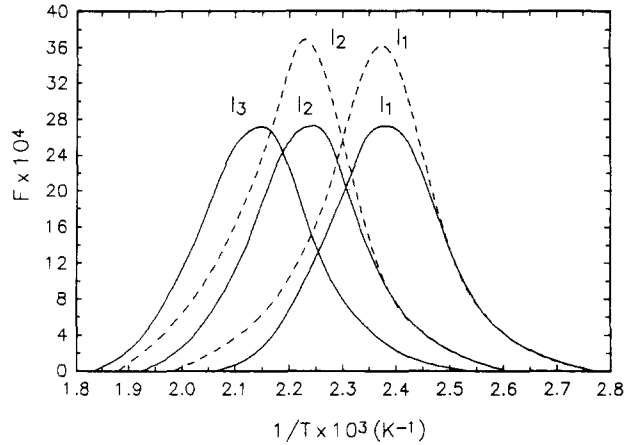

Fig. 1. Internal friction vs. the reciprocal of the temperature measured at three moments of inertia (I_1, I_2, I_3) in the specimen with 550 at.ppm O and two moments of inertia (I_1, I_2) in the specimen with 770 at.ppm O.
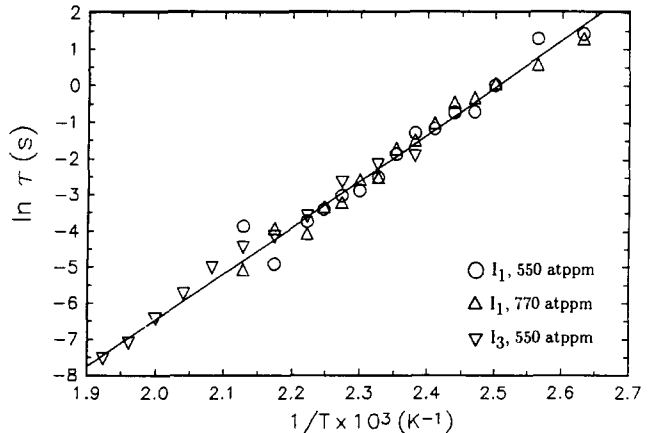

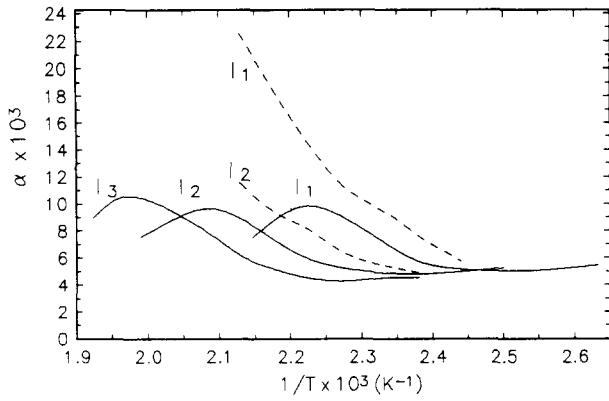Fig. 2. Relationship between $\ln \tau$ and $1/T$.

Fig. 3. The values of α against the reciprocal of temperature for the peaks shown in Fig. 1.

of the moment of inertia (or the frequency). In fact, once τ is known, it is possible to calculate α for each peak of Fig. 1, by using eqn. (4). The values of α obtained for the various peaks are indicated in Fig. 3, plotted against the reciprocal of the temperature. It is clear that α not only depends on the temperature but also on the moment of inertia of the pendulum or, equivalently, on the frequency. This indicates that the internal friction peaks shown in Fig. 1 are not of the Debye type and a distribution function of relaxation times is involved in the Snoek relaxation.

4. Discussion

It is generally stated in the literature that for oxygen concentrations lower than 10^3 at.ppm the Snoek relaxation in niobium can be described with high accuracy in terms of a single relaxation time. As shown in the previous section, this is not the case even for oxygen contents as low as 550 at.ppm.

Diehl et al. [8] have shown that the halfwidth of the log–normal distribution of relaxation times, as obtained using the procedure described by Nowick and Berry [9], decreases with the oxygen concentration in niobium, being practically zero below about 1000 at.ppm. Furthermore, according to Haneczok and Weller [10], the Snoek peak in niobium is of the Debye type at an oxygen concentration of 70 at.ppm. This conclusion was reached, however, by using only one internal friction peak and the temperature dependence of the oscillation frequency. In addition, it was assumed by the authors that the relaxation strength decreases linearly with the reciprocal of the absolute temperature. In a more recent publication Weller et al. [11] have reported a value of 0.41 for the halfwidth of the log–normal distribution of relaxation times for an oxygen concentration of 1200 at.ppm. They also reported the following parameters for the Snoek peak:

$$\tau_0 = 2.89 \times 10^{-15}\ \text{s}, \qquad H = 1.14\ \text{eV} \qquad (10)$$

The value for H is very close to that given by eqn. (9).

It should be pointed out that a completely different approach used to fit the experimental internal friction data in an Nb–0.6at.%O alloy [2] leads to the conclusion that a distribution in both τ_0 and H must be involved in the relaxation process.

On assuming that the distribution of relaxation times is log–normal, it is possible to obtain the parameters of the distribution and the relaxation strength from the measured F and ω vs. T curves by using a procedure which will be described briefly [12]. The partial derivative of the oscillation frequency with respect to the moment of inertia can be measured at each temperature. This derivative is given by

$$2\left.\frac{\partial \ln \omega}{\partial I}\right|_{(1/T)} + \frac{1}{I}$$

$$= \frac{\partial \ln}{\partial I}\left(1 + \delta \int_{-\infty}^{\infty} \frac{\psi_\epsilon(\ln \tau)\omega^2\tau^2}{1+\omega^2\tau^2}\, \mathrm{d}\ln \tau\right)_{(1/T)} \qquad (11)$$

This equation solved for δ gives the relaxation strength as a function of temperature in terms of the parameters of the distribution, i.e. $\delta(\beta, \tau_m, T)$, where β is the halfwidth and τ_m is the mean value of the relaxation time for the distribution function Ψ_ϵ at constant strain. On taking into account the relationship between Ψ_ϵ and Ψ_I, which is the distribution function associated with the internal friction [2], it is possible to obtain β and δ separately as a function of temperature. The results obtained for the peaks shown in Fig. 1 are indicated in Figs. 4 and 5. Even with some dispersion in the calculated values, β decreases almost linearly with temperature, with a similar behaviour for both oxygen concentrations, as illustrated in Fig. 4. The temperature dependence of δ is influenced by the oxygen content, as illustrated in Fig. 5. β varies with temperature
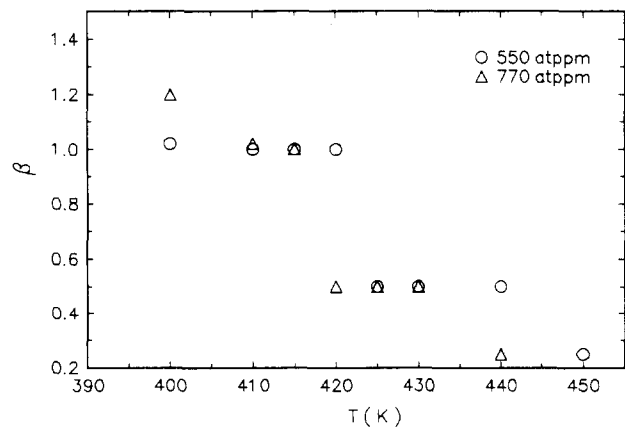

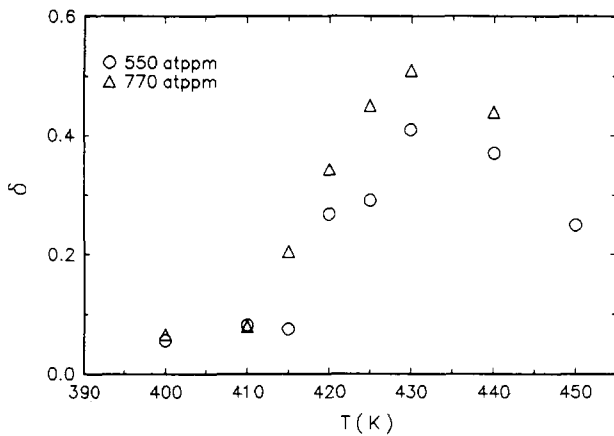Fig. 4. β against temperature for the peaks shown in Fig. 1.

Fig. 5. δ against temperature for the peaks shown in Fig. 1.

in the way suggested by Nowick and Berry [9], while the temperature dependence of δ is more complicated than simply a Curie–Weiss-type behaviour. It should be pointed out that the procedure described does not make any *a priori* assumption about the temperature dependence of the parameters and δ and β are evaluated at each temperature through the internal friction peak. On using the method proposed by Nowick and Berry [9], which is generally used in the literature, only one value of β is assigned to each internal friction peak. This might not be correct if β depends on the temperature. We do not have a physical model to explain the temperature dependence of β shown in Fig. 5. Okamoto [13] has studied the mobility of oxygen in niobium by means of a strain relaxation technique. The relaxation curve has been fitted by a summed exponential model and five relaxation processes were present in an Nb–0.7at.%O alloy. The relaxation strength for the first process was found to increase with temperature. This relaxation process was attributed to the jump of single oxygen atoms. Haneczok *et al.* [14] have assumed a model including long-range interactions of interstitials in niobium in order to explain the Snoek relaxation. It is clear that the interpretation of the results is still controversial and further work is needed.

A detailed discussion of these results will be given in another publication [12], but it should be pointed out that an attempt to fit the peaks of Fig. 1 with a log–normal distribution with β and δ given by Figs. 4 and 5 does not give adequate results near the tails of these peaks.

5. Conclusions

Internal friction measurements of the Snoek relaxation in Nb–O alloys with low oxygen contents using a torsion pendulum with variable moment of inertia have shown that the relaxation process is not controlled by a single relaxation time. The parameter of the log–normal distribution function, assumed to describe the experimental data, and the relaxation strength have been obtained as a function of temperature and oxygen content.

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