INTEGRATED QUASI-ELASTIC NEUTRON SCATTERING INTENSITY OF HYDROGEN IN NIOBIUM

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The integrated quasi-elastic neutron scattering intensity of $NbH_{0.13}$ has been measured at 235°C over a large momentum transfer range ($0.6 < K < 7$ Å^{-1}). In the high temperature phase a deviation from a simple Debye-Waller factor governed intensity dependence has been observed.

Most of the quasi-elastic neutron scattering measurements performed so far on hydrogen in metals [1] has been analyzed according to a simple jump diffusion model [2], in which it is assumed, that the mean residence time τ_0 of the hydrogen at any interstitial site is large compared to the jump time τ_1, leading to an intensity dependence $\sim\exp(-u^2K^2)$ (u^2 is the mean square amplitude of the hydrogen and $\hbar K$ the momentum transfer). Recently it has been supposed [3, 4] that in the high temperature phase τ_1 could be of the same order of magnitude as τ_0; this should give rise to a deviation from the Debye-Waller factor determined intensity dependence.

To look for such a deviation we have measured the integrated quasi-elastic scattering intensity I (area below the quasi-elastic line) of $NbH_{0.13}$ at 235°C in the α-phase and at 20°C in the mixed ($\alpha+\beta$)-phase over a large momentum transfer range $0.6 < K < 7$ Å^{-1}. In the mixed phase the hydrogen diffusion constant is about 100 times smaller [5] than in the α-phase [6]. On the other hand in both phases the interstices occupied by the hydrogen seem to be the same (tetrahedral sites) [4, 5], the local and optical mode energies respectively are nearly equal [7, 8] and in the β-phase the host-lattice structure is only slightly distored [9].

The samples used were cylindrical single crystals. The hydrogen loading technique has been described earlier [4]. The measurements have been performed with a rotating crystal spectrometer [10] using incident energies (E_0) 0.0143 eV and 0.057 eV ($\Delta E/E_0$ = 7.5%, scattering angle divergency $\Delta(2\vartheta) = 1°$). The pure hydrogen quasi-elastic scattering intensity has been determined by the difference of the spectra of a loaded and an unloaded sample. In order to avoid errors due to long background variations the samples were interchanged every half hour. The normalized difference spectra thus obtained have been corrected for constant background and counter efficiency and then converted into an energy scale. The area of the quasi-elastic line has been determined by the use of a planimeter.

Fig. 1 shows the final results in a logarithic plot of the intensity I versus K^2 or both temperatures. The indicated error bars are mainly due to background subtraction errors and only slightly to counting statistics. The circles and the triangles respectively are the results of the measurement at $E_0 = 0.0143$ eV and $E_0 = 0.057$ eV after adjusted to give one smoothed curve. A significant deviation from the simple Debye-Waller factor determined intensity dependence is observed in the high temperature phase for $K^2 < 6$ Å^{-2}. In the remaining region, and in the entire measured K range of the mixed phase, the intensity fits a Debye-Waller factor dependence corresponding to a mean square amplitude of $u^2 = 0.016 \pm 0.005$ Å^2 for 235°C and $u^2 = 0.020 \pm 0.005$ Å^2 for 20°C. Earlier u^2 has been found to be equal to: 0.18 Å^2 for $NbH_{0.33}$ at 308°C [11] in the range $0.8 < K < 2.2$ Å^{-1}, 0.08 Å^2 for $NbH_{0.14}$ at 250°C [12] in the range $0.5 < K < 2.1$ Å^{-1}, 0.05 Å^2 for $NbH_{0.09}$ at 235°C [13] in the range $2.0 < K < 4.0$ Å^{-1}. Approximately the same values could be extracted from our measurements in the correspondingly limited K ranges.

Considering finite jump times the quasi-elastic scat-

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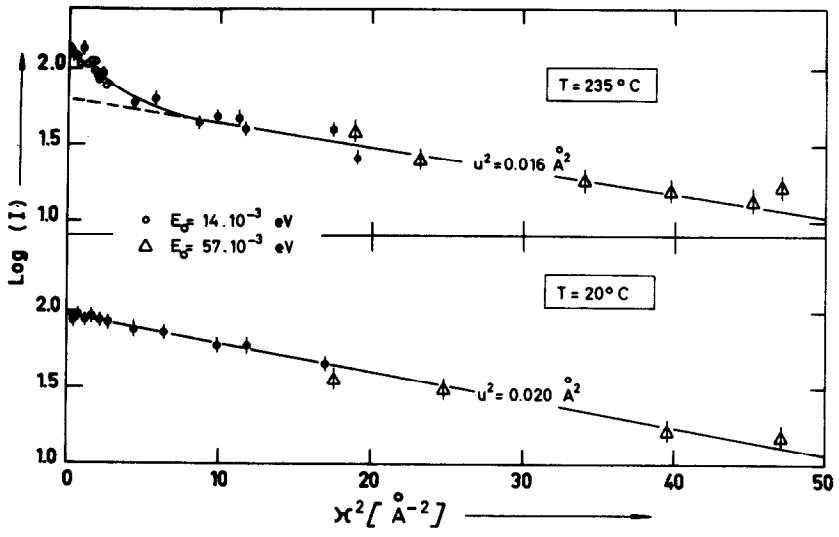

Fig. 1. Logarithm of the integrated quasi-elastic neutron scattering intensity versus K^2 for 235°C and 20°C. The values given by the triangle have been fitted to the curve given by the circles.

tering intensity is roughly given [14] by two terms $I \sim (A + B)$, where $A = \exp(-u_R^2 K^2) \cdot \tau_0/(\tau_0 + \tau_1)$ represents the intensity due to the hydrogen atoms trapped at interstitial sites (mean square amplitude u_R^2) and $B = F(K) \cdot \tau_1/(\tau_0 + \tau_1)$ is the intensity due to the hydrogen atoms in the jump phase. Suppose the existence of a diffusion band $F(K)$ represents a time averaged structure factor of the hydrogen wave packet in this band, having a limited life time τ_1. Classically we expect $F(K) = 1$ for free jumping particles or $F(K) = \exp(-u_j^2 \cdot K^2)$ for particles which are trapped in an intermediate potential in between the interstitial sites, performing there oscillations with a mean square amplitude u_j^2 as supposed in [16].

In the low temperature phase we expect $\tau_1 \ll \tau_0$, therefore it is mainly the A term which contributes to the total intensity. Actually the experimental results show the expected simple Debye-Waller factor dependence with $u_R^2 = 0.02$ Å^2. Using the Einstein oscillator approximation and taking for the oscillator frequency a value of $1.8 \times 10^{14} s^{-1}$ (lowest measured optical mode frequency in the β-phase [8] we find that $u^2 = 0.017$ Å^2. We assume that in the high temperature phase the intensity dependence for $K^2 > 6$ Å^{-2} is also mainly due to the A term, whilst for $K < 6$ Å^{-2} the B term begins to have an influence. Then we can separate the B term contribution by subtracting the intensity, which is expected to be due to the A term in the small K range, from the total measured intensity. Fitting the resulting curve again to a Debye-Waller factor intensity dependence we get $u_j^2 \approx 0.4$ Å^2. From the ratio of the intensities extrapolated to $K \to 0$ we obtain $\tau_1/\tau_0 = 0.45$. Both values are exceptionally large, taking into account that the distance to the next interstitial site is only 1.17 Å and that J_0 has been determined to be $\approx 10^{-12}$s [4]. This might be evidence for the existence either of the previously assumed diffusion band [15, 17] or of an intermediate state [16]. A free jumping particle model can be excluded.

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