Anisotropic Diffusion of Hydrogen in Niobium Single Crystals

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Quasielastic neutron scattering experiments on hydrogen in niobium single crystals have been performed in the $\vec{K}$ range from 1.7 to 4.3 $Å^{-1}$ in the three directions [100], [110], and [111]. The experimental width functions show a significant anisotropy. The results are not in agreement with the theoretical half-widths calculated for tetrahedral and octahedral positions of the hydrogen.

In the last few years measurements[1-4] have been reported about quasielastic scattering of neutrons on hydrogen in transition metals. By measuring the quasielastic broadening as a function of the momentum transfer and the temperature, the microscopic diffusion process can be investigated. So far these experiments have been performed with polycrystalline samples. The use of single crystals has the advantage that the physical interpretation is simpler because the momentum transfer $\vec{K}$ relative to the lattice directions is well defined. In the case of PdH,[3] which has a fcc lattice, the sites have been determined. For NbH[3] and VH,[4] which have a bcc lattice, the experimental results obtained so far with $\vec{K}$ values smaller than 2.2 $Å^{-1}$ did not lead to definite conclusions about the lattice sites and the microscopic diffusion process. However, the diffusion constant and the activation energy are known from both relaxation time measurements[5] and quasielastic scattering[2,3] of neutrons at small $\vec{K}$ values.

The experiments have been performed with the Ispra rotating crystal spectrometer.[6] This instrument allowed measurements in the $\vec{K}$ range from 1.7 to 4.3 $Å^{-1}$. The broadening has been determined with $\vec{K}$ parallel to the [100], [110], and [111] directions of the lattice. The sample consisted of two single crystals with dimensions $2\times1.2\times0.4$ cm^3. The hydrogen content was 9 at. %. The effect of the multiple scattering could be neglected, because the transmission of the sample at 237°C, which was measured, was 0.94. The measurements have been performed at 237° C, where a conveniently measurable broadening has been observed. The energy resolution ΔE was 0.7 meV. The main experimental difficulty was the selection of $\vec{K}$ values at which no coherent contribution appeared in the elastic spectrum of niobium. The main error of the results is connected with the background subtraction, whereas the contribution of the inelastic scattering is of less importance than in the case of liquids with the same diffusion constant. We determined the physical widths by folding[7] the resolution function obtained by a vanadium standard with Lorentzians and comparing them with the experimental results. The results are shown in Fig. 1.

Theories for the calculation of the diffusive broadening of quasielastic scattering by interstitials forming a non-Bravais lattice have been developed by Blässer and Peretti[8] and Gissler and Rother,[9] using different mathematical methods. Calculations have been performed for different lattice positions of the interstitials. In the case of a non-Bravais lattice the scattering function is a sum of Lorentzians, the number of which depends on the number of nonequivalent sites.

Gissler and Rother[9] have calculated the half width at half-maximum for the theoretical scattering function, which is generally not a Lorentzian, for tetrahedral and octahedral sites and for single-crystal samples.

Their results cannot be compared directly with the experiment. Therefore, we have folded the different Lorentzians, calculated by the theory of

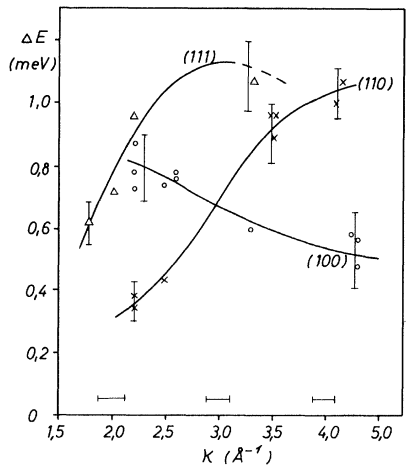

FIG. 1. Physical width functions for the three axes obtained by quasielastic neutron scattering on hydrogen in a Nb single crystal.

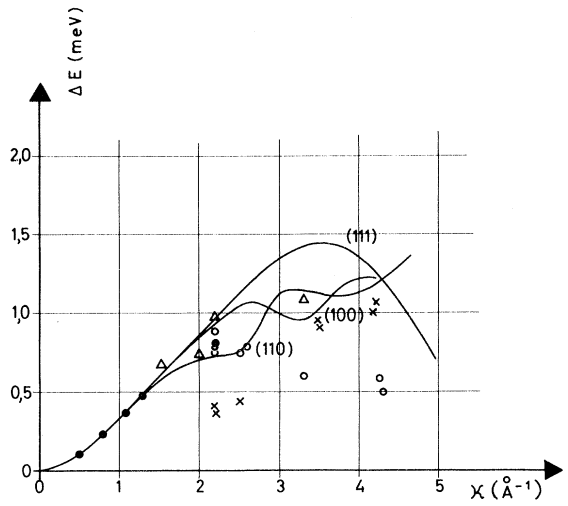

FIG. 2. Theoretical width functions, solid lines, in the case of tetrahedral sites for the three axes, determined from a sum of the Lorentzians calculated by the Blässer and Peretti theory (Ref. 8), each folded with the resolution function. Physical widths for the [100], [110], and [111] directions: open circles, crosses, and triangles, respectively. Polycrystalline results (Refs. 2, 11), closed circles. The theoretical curves have been fitted to polycrystalline results for small $\vec{K}$ values.

Blässer and Peretti,[8] with the resolution function. The results, shown in Fig. 2 with the assumption of tetrahedral positions, are not in agreement with the experiment, which shows a much stronger anisotropy.

Recently the same calculations have been performed for the case of octahedral sites by Gissler[10] and Blaesser.[11] Their results are also in disagreement with the experimental values. A value of 0.05 ± 0.01 Å^2 has been found for the mean square amplitude of vibration, u^2, for each of the three directions.

From the results it can be concluded that the hydrogen in Nb is not located at tetrahedral sites, as assumed so far. Furthermore the results also allow us to exclude octahedral sites.

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