JETP Letters, Vol. 76, No. 5, 2002, pp. 318–320. Translated from Pis'ma v Zhurnal Éksperimental'noĭ i Teoreticheskoĭ Fiziki, Vol. 76, No. 5, 2002, pp. 377–379.
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Hydrogen Tunneling Modes in α-Mn Suppressed by Elastic Stresses

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Received August 6, 2002

Behavior of the tunneling mode of hydrogen in $MnH_{0.04}$ and $MnH_{0.07}$ under high pressures in sapphire anvils was studied by the method of incoherent inelastic neutron scattering (INS). It is established that the INS peak corresponding to the hydrogen tunneling in a double-well potential disappears at a pressure of 0.8 GPa in a quasi-hydrostatic regime, while being retained without visible changes under pure hydrostatic conditions. An analogous, albeit weaker, suppression of the tunneling mode takes place upon grinding of a freshly prepared sample. The effect of suppression of the hydrogen tunneling modes by applied inhomogeneous elastic stresses is explained by a shift of the energy levels in the adjacent wells caused by the static displacements. © *2002 MAIK "Nauka/Interperiodica".*

PACS numbers: 62.50.+p; 61.12.Ex

Recent experiments [1–3] on the neutron diffraction and inelastic neutron scattering (INS) from solid solutions of hydrogen in α-Mn revealed the effect of hydrogen tunneling at relatively high temperatures (up to about 90 K). The effect was manifested by the appearance of a temperature-dependent intense narrow peak at 6.3 meV in the INS spectra, which was explained by splitting of the ground vibrational state as a result of the tunneling of hydrogen atoms between adjacent closely spaced 12e positions. The energy of the observed tunneling modes was about 30 times that for the other quantum systems [4, 5].

The nature of the observed effect is probably related to the fact that hydrogen atoms incorporated into complex Bravais lattices (such as those of α- and β-Mn) occur in positions spaced much more closely as compared to the case of hydrogen solutions in other metals possessing simple lattices. For example, the neutron diffraction data [2] showed that hydrogen atoms in α-Mn randomly occupy 12e dumb-bell positions in the space group *I*43*m*, the distance between which (0.68 Å) is much shorter than that typical of the hydrogen atoms dissolved in bcc metals (2.2 Å). It was natural to suggest that a decrease in volume, leading to a reduction of the distances between possible positions of hydrogen atoms and to a change in the shape of the double-well potential, may significantly influence the tunneling of hydrogen. In connection with this, we studied the effect of high pressures on the effect of hydrogen tunneling in α-Mn by method of incoherent INS.

The experiments were performed on the $MnH_{0.04}$ and $MnH_{0.07}$ samples prepared as described previously [6]. The powder of α-Mn was saturated with hydrogen (generated via decomposition of aluminum hydride) in a high-pressure chamber. The hydrogenation process was conducted at a pressure of 0.85 GPa and a temperature of 623 K for 4 h, followed by rapid cooling to room temperature. The content of dissolved hydrogen was determined by hot extraction into a calibrated volume.

The neutron scattering measurements were performed in a temperature interval from 15 to 100 K on a DN-12 spectrometer operating in the INS mode [7] using an IBR-2 reactor of the Laboratory of Neutron Physics (JINR). For this purpose, a ring-shaped pyrolytic graphite analyzer was placed between the samples and a ring detector composed of 16 ^{3}He counters of the SNM-31 type, so that the scattering angle was $2\theta = 90°$ and the energy analyzed was 14.9 meV.

High pressures were applied to the samples placed into special cells with sapphire anvils [8]. Samples with a volume of ~2 mm^3 were loaded at a pressure of up to 20 kbar in a quasi-hydrostatic regime. Purely hydrostatic conditions were created by filling anvils with a special fluid (Fluorinert). Samples in a high-pressure chamber were cooled using a special cryogenic refrigerator.

The INS spectra of freshly prepared samples (Fig. 1a) showed that a decrease in temperature below 100 K leads to the appearance of a sharp resonance

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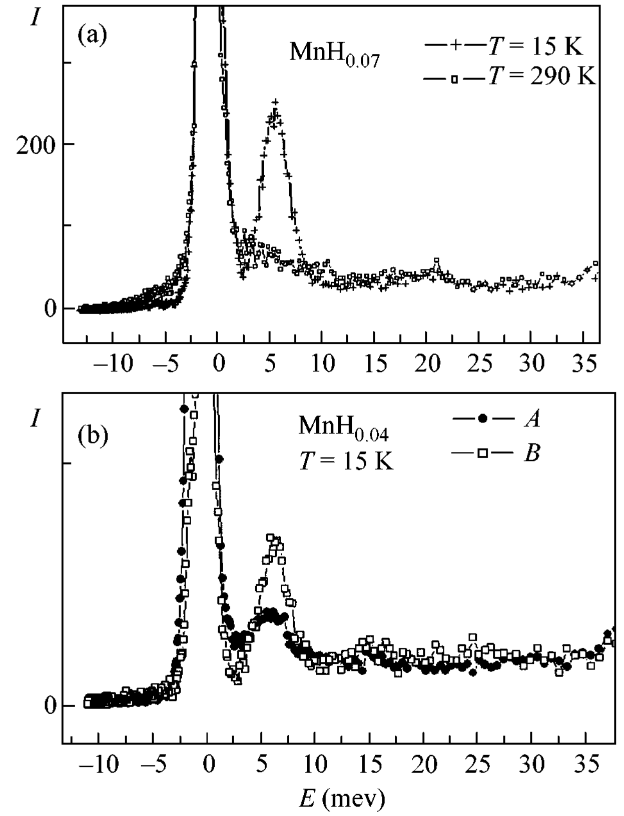

Fig. 1. INS spectra measured on a DN-12 spectrometer (a) for $MnH_{0.07}$ at different temperatures and (b) for freshly prepared and triturated $MnH_{0.04}$ samples.

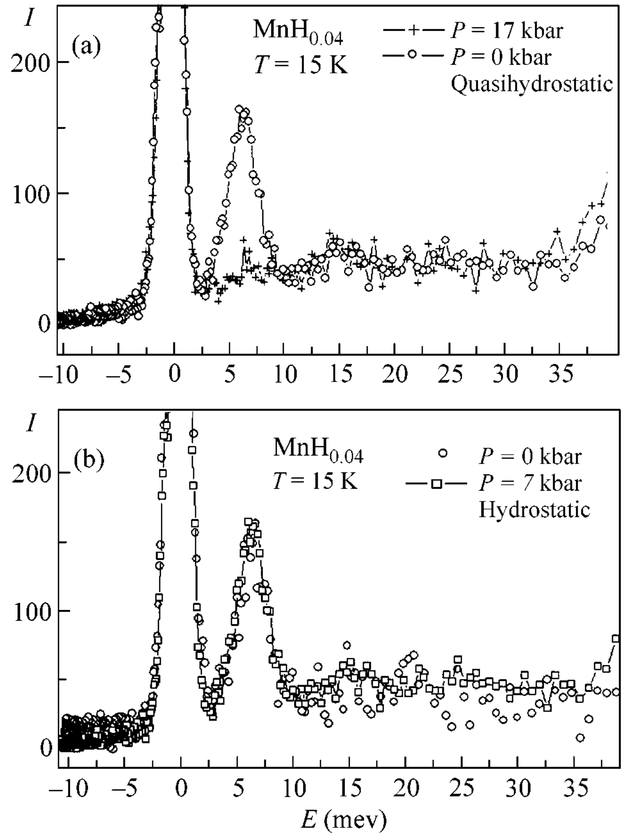

Fig. 2. INS spectra measured on $MnH_{0.04}$ samples loaded by different pressures in sapphire anvils under (a) quasi-hydrostatic and (b) purely hydrostatic conditions.

peak at 6.4 meV, which agrees with the previous results [3] and corresponds to the tunneling motion of hydrogen atoms in the double-well potential. The peak amplitude significantly decreases upon dry or wet (ethanol) trituration of the samples (Fig. 1b). Upon loading in a quasi-hydrostatic mode in sapphire anvils, the INS spectrum exhibits a dramatic change: the peak at 6.4 meV disappears already at a pressure of about 8 kbar and is restored neither at higher pressures (17 kbar) nor upon unloading (Fig. 2a). Since the phenomenon is observed in both $MnH_{0.04}$ and $MnH_{0.07}$ samples, the effect of pressure is independent of (or weakly dependent on) the hydrogen concentration.

The above results suggest that the effect of suppression of the hydrogen tunneling states is related to the residual elastic stresses (or defects) arising in the samples as a result of nonhydrostatic loading or grinding, while the contribution of homogeneous compression is small. In order to verify this assumption, we conducted experiments under hydrostatic conditions, using freshly prepared samples loaded in anvils filled with Fluorinert. In this case, no significant changes in the INS peak at 6.4 meV were observed: the intensity and the width and position of the peak remained virtually the same upon hydrostatic loading (Fig. 2b).

Thus, changes in intensity of the INS peak related to hydrogen tunneling are caused by the local elastic stresses in the sample, either induced by inhomogeneous pressure or produced by grinding, rather than by applied pressure as such.

The probability of tunneling in a defect-free crystal is determined as

$$W \approx \Delta^2/\omega,$$

where Δ is the tunneling bandwidth and ω is the characteristic frequency of the dynamic displacements caused by the interaction with lattice vibrations [9, 10]. The probability of tunneling in a crystal strained by elastic stresses must be significantly decreased by a shift δ between the energy levels:

$$W \approx \Delta^2 \frac{\omega}{\omega^2 + \delta^2}.$$

In the absence of local stresses, a shift of levels arises due to the dynamic displacements and determines the temperature dependence of the tunneling probability. In our experiments, the shift of levels is determined by static (rather than dynamic) displacements. The large magnitude of this shift is explained by the elastic moduli of the metal matrix being much higher as compared to those of a quantum crystal [11]. Analogous manifestations of the tunneling modes suppressed by local elastic stresses should probably be observed in other quantum systems as well, albeit being less pronounced in cases of lower elastic moduli.

The authors are grateful to L.A. Maksimov for fruitful discussions and valuable remarks, and to Yu.M. Kagan and E.G. Ponyatovskiĭ for discussion of the results.

This study was supported by the Russian Foundation for Basic Research (project nos. 00-02-17077 and 99-02-17299) and by the Federal Program "Leading Scientific Schools" (project no. 00-15-96712).

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Translated by P. Pozdeev