

Lattice dynamics of high-pressure hydrides of the group VI–VIII transition metals

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Monohydrides of the group VI–VIII transition metals Cr, Mn, Fe, Co, Ni, Mo, Rh and Pd were synthesised under high hydrogen pressures at ISSP RAS and studied by inelastic neutron scattering (INS) at ILL. The hydrides have close-packed metal sublattices with hcp, fcc or dhcp structure, in which hydrogen atoms occupy octahedral interstitial positions. The fundamental band of optical hydrogen vibrations in these hydrides consists of a strong peak with a broad shoulder towards higher energies. Based on results for palladium hydride [1], the peak is usually ascribed to nearly non-dispersive transverse optical modes, while the shoulder is assumed to arise from longitudinal optical modes, which show significant dispersion due to long-range repulsive H–H interaction. The energy of the main optical H peak exhibits a strong monotonous increase as a function of the hydrogen–metal distance, R , in the hydrides of both 3d- and 4d-metals. The increase in R by itself should imply weakening interatomic interactions, and the observed opposite effect thus indicates a significant increase in the hydrogen–metal interaction with decreasing atomic number of the host metal, which outweighs the influence of the accompanying R decrease in the hydrides. This effect agrees with predictions of first-principles calculations [2] that the reduction of the number of valence electrons from right to left along the transition metal rows in the periodic table should lead to a steepening of the potential well for interstitial hydrogen due to a less efficient screening of the ionic core charges.

Our INS studies of textured foils of *fcc* PdH [3] and NiH [4] and single-crystalline PdH_{0.85} [5] demonstrated that, despite the cubic symmetry of the octahedral hydrogen positions in these hydrides, the second and higher bands of optical H vibrations are strongly anisotropic due to an anisotropy of the potential well for H atoms. Earlier, no significant anisotropy had been observed in the INS spectra of palladium hydride or any other hydride with cubic symmetry of hydrogen positions. The observed behaviour of the spectra at higher energies can be qualitatively described by using *ab-initio* calculations [6,7].

Analogous calculations predicted that the second optical band in PdD [6] and NiD [7] should also exhibit a strong anisotropy similar to that in PdH and NiH. However, our recent INS studies of textured foils of *fcc* PdD [8] revealed no anisotropy in the spectrum of deuterium vibrations at energy transfers below 110 meV. In particular, the peak at $\omega_2^D \approx 72$ meV in the second D band in PdD showed no anisotropy in contrast to the H peak at $\omega_2^H \approx 112$ meV in PdH. This indicates that the potential well for H and D atoms in palladium is nearly isotropic at energies up to $5/4\omega_2^D \approx 90$ meV, counted from the bottom of the well, and becomes strongly anisotropic only at energies exceeding $5/4\omega_2^H \approx 140$ meV.

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