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Dynamical effects in solid hydrogen and hydrogen-deuterium mixtures at elevated pressures.

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We employed molecular dynamics simulations together with some novel theoretical techniques to calculate finite temperature Raman spectra in both hydrogen (H) and hydrogen-deuterium (HD) mixtures. By conditioning the simulations with the experimental data, we have discovered the true nature of Phase IV in solid hydrogen. X-ray and neutron diffraction measurements are virtually impossible in current state of the art experiments. The experimental investigation, thus, relies mainly on spectroscopic methods which provide minimal data about crystallography. Extensive work done so far [1] has led to an impressive amount of Raman and IR data up to pressures of almost 400 GPa. By carefully investigating the positions and widths of the peaks, they discovered phase IV of solid hydrogen and mapped out the melting curve up to very high pressures. Phase IV is only stable at elevated temperatures and therefore static relaxation calculations cannot reproduce the correct crystal structure. Previous theoretical work has predicted a class of exotic crystal structures with alternating layers of different character: B (well-defined molecules) and G (poorly-defined molecules) [2] that could qualitatively describe the Raman signal. By means of MD simulations we show that at high temperature, the Pc structure acquires higher symmetry which is sufficient to reproduce the exact position of the Raman peaks. It had been suggested that in a mixed hydrogen-deuterium alloy, the heavier atoms would segregate in the B layers which would decrease the overall zero-point energy and stabilise the Pc structure. Joining efforts with the experimental team we thoroughly investigated this problem. We found no evidence of isotopic layer segregation, but our theoretical model correctly described the Raman spectra found experimentally. We concluded that the phase boundaries in HD mixtures are slightly offset which we attribute to the inhomogeneous mass effect on the temperature of the broken symmetries, but the general behaviour is similar to that of solid hydrogen. We uncovered a new effect in HD, namely mass induced phonon localization. Matching experimental and theoretical results in this respect are yet another validation of the crystallographic conformation of phase IV.

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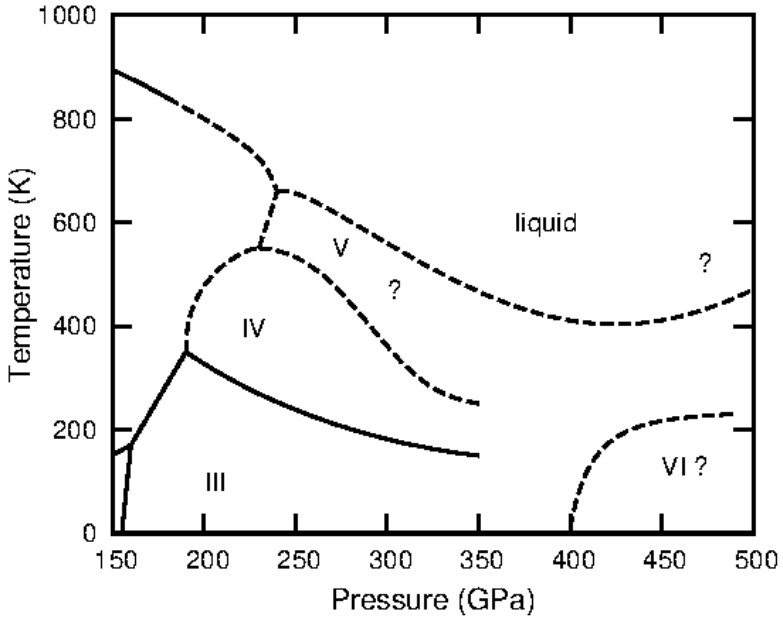

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