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Effect of Collective Oscillations on Low-Energy Thermal-Neutron Scattering and Transport in Liquid Hydrogen

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A thermal-neutron inelastic scattering kernel based on recently observed collective motion in liquid hydrogen, taken to be of quasicrystalline nature with an appropriate chemical binding energy, is found to explain the experimental pulsed neutron and steady-state spectra studies at 20 K. Thus the collective motion plays a very significant role in the transport of low-energy thermal neutrons in liquid hydrogen.

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In a recent study Carneiro, Nielsen, and McTague[1] have reported the presence of collective motion in liquid parahydrogen at 14.7 K observed by them using coherent neutron inelastic scattering. A distinguished feature of their measurement *vis à vis* other measurements[2,3] on monatomic coherent liquids is the presence of two peaks in the scattering law corresponding to the transverse and longitudinal modes similar to what one expects in a polycrystal. The significance of the presence of collective motions in liquid parahydrogen particularly for low-energy thermal-neutron inelastic scattering, and hence transport studies, is shown by the experiment itself. If the collective motions were absent, or were of minor importance, the scattering law would not have contained the low-energy dynamical structure. In another similar experiment on solid hydrogen performed at 5.4 K, Nielsen and Carneiro[4] have found that the phonon dispersion curves are similar to those of continuum model of a solid. Even the measured values of mean square displacements in C direction and the basal plane are almost equal, i.e., the mean square displacement of a hydrogen molecule is isotropic even in hydrogen crystal. Thus, it is evident that the collective motion is not only present in liquid hydrogen, but that it can be approximated rather well by the collective motion of a continuous medium. This conclusion is consistent with the inference of Carneiro, Nielsen, and McTague, based on their experimental results on liquid hydrogen, that in several respects the dynamics of liquid hydrogen is solidlike. We, therefore, suggest that the effect of collective motion on low-energy thermal-neutron scattering and hence transport from liquid hydrogen may be like that from a continuous or a quasicrystalline medium, with an appropriate chemical binding energy represented by a characteristic Debye temperature, Θ_D. Such a model for the dynamics of liquid hydrogen has, so far, not been used for thermal-neutron scattering and transport studies.

There have been several attempts[5-8] to explain the total neutron-scattering cross sections of thermal neutrons from liquid hydrogen. But all these attempts, though take into account the scattering of neutrons from energetic molecular

modes, i.e., rotational and vibrational modes, neglect the chemical binding energy, which is the most important characteristic of liquid state as opposed to the gaseous state, and hence ignore the scattering of neutrons from the collective motions of liquid hydrogen. The collective oscillations of liquid hydrogen give rise to dense low-energetic modes through which neutrons can lose energy, and will therefore, play a very important role in the cold-neutron scattering,[9] and hence transport[9] also, as evidenced by the recent experimental results.[1] Since the earlier thermal-neutron scattering kernels of liquid hydrogen treat liquid hydrogen as if it were in gaseous state, these are no longer acceptable, as they are not in accordance with the recent experimental results.

In this Letter, we attempt to show that it is possible to construct a realistic thermal-neutron inelastic scattering kernel for liquid hydrogen which takes into account the presence of collective modes in liquid hydrogen in the continuum approximation, with a cutoff energy equal to $k_B\Theta_D$. For scattering processes we consider up to two phonon exchanges. As the temperature at which liquid hydrogen is used for neutron scattering and transport is never more than 21 K, the neutrons which affect these studies are not very energetic and therefore, we can neglect the contribution of rotational and intramolecular vibrational modes in the estimation of inelastic scattering. The minimum energy of neutrons which can excite a parahydrogen molecule in liquid state to its first rotational excited state is[9] 0.015 eV, which is quite large compared to energies of most of the nearly thermalized neutrons at 20 K. The steady-state neutron spectrum at 20 K has a peak at 2×10^{-3} eV around which (i.e., 10^{-4} to 8×10^{-3} eV) most of the neutrons are present. [It may be noted that in liquid state, because of the presence of chemical binding energy, the recoil energy of a hydrogen molecule in the exchange process is negligible, as is also shown by the experimental results on total scattering cross sections of liquid parahydrogen at different temperatures.[10-12] This would not be so if chemical binding energy in liquid hydrogen were negligible.] Further, since we are studying the effect of collective oscillations at a low temperature (< 21 K) and at low neutron energies (< 0.015 eV), the suggested scattering kernel will not differentiate between orthohydrogen and parahydrogen. Though their nuclear configurations are different, their electronic behaviors are the same,[5] and therefore, the chemical binding energy in the two states will be the same. [This is also indicated by the experimental results of steady-state neutron spectra from different types of hydrogen at 20 K. The spectra from parahydrogen and 75% ortho and 25% para mixture are not very much different.[9,13]] The scattering kernel can therefore be applied with great ease to study the problem of low-energy thermal-neutron transport in ortho-para mixtures also.

We consider now the following two experiments, involving the transport of neutrons for which the experimental results are also available. We consider first the decay of a neutron pulse in liquid hydrogen for its various assembly sizes. The experimental results[14] on liquid parahydrogen are in a small buckling range ($B^2 \leq 0.1$ cm^{-2}) and on ortho-para mixture in a larger buckling range ($B^2 \leq 0.8$ cm^{-2}). The calculation of asymptotic decay constants, $\lambda_c = \lambda_{n=0}$, of a neutron pulse in an assembly reduces to solving the following multigroup Boltzmann transport equation[15]

$$\sum_j P_{ij} N_n(V_j) = \lambda_n N_n(V_i), \tag{1}$$

where

$$P_{ij} = -v_0 V_j \Sigma^s(V_j \to V_i) dV_j + v_0[V_i\Sigma_i^a + V_i\Sigma_i^s + V_i D_i B^2].$$

Here V_i and V_j are the velocities of i and j groups, respectively, expressed in the units of velocity $v_0 = (2k_BT/m)^{1/2}$ where T is the temperature of the assembly, k_B is the Boltzmann constant and m is the mass of neutron; Σ_i^a is the macroscopic absorption cross section for neutrons of energy group i; B^2 is the geometric buckling[16] and is related to the dimensions of the assembly (for a cubic assembly of side a, $B^2 \simeq 3\pi^2/a^2$); $D_i = [3(1 - \langle\cos\theta\rangle)(\Sigma_{i,T}^s + \Sigma_i^a)]^{-1}$, $\Sigma_{i,T}^s$ being the total macroscopic scattering cross section and both are different for orthohydrogen and parahydrogen; for a given ortho-para hydrogen mixture having X% orthohydrogen and the rest parahydrogen $\Sigma_{i,T}^s = [\Sigma_{i,T}^s(o\text{-}H_2)\times(X) + \Sigma_{i,T}^s(p\text{-}H_2)\times(100-X)]$; $\Sigma^s(V_j \to V_i)$ is the macroscopic neutron inelastic scattering kernel and is given by[17]

$$\Sigma^s(V_j \to V_i) = \rho_n[\sigma_1(V_j \to V_i) + \sigma_2(V_j \to V_i)],$$

where ρ_n is the number of scattering centers per unit volume, $\sigma_1(V_j \to V_i)$ and $\sigma_2(V_j \to V_i)$ are the one- and two-phonon kernels, respectively. $\langle\cos\theta\rangle$ is the average of the cosine of the scattering angle of the neutron in the laboratory system.

In Fig. 1(a), the calculated results of asymptotic decay modes for liquid 47% parahydrogen and 53% orthohydrogen mixture assemblies of various sizes are plotted at 20 K along with the experimental results.[14] We calculated λ_0 with various values of Θ_D using one- plus two-phonon scattering kernels and found best agreement with experimental values for $\Theta_D = 80$ K only. [The calculated values given by one-phonon scattering kernel are lower than the experimental values particularly for large B^2. The values given by gas kernel with mass of the scatterer as 2 are much larger than the experimental values. These values are also shown in Fig. 1(a).] This value of $\Theta_D = 80$ K is in agreement with the oft quoted[7,13] value of $\Theta_D \sim 0.007$ eV for liquid hydrogen and is also physically consistent, as it is lower than the $\Theta_D = 117$ K for solid hydrogen[4,18] at 0 °K and $\Theta_D = 103$ K for solid hydrogen at melting point.[4] Using the value of $\Theta_D = 80$ K for liquid hydrogen we calculated the mean square displacement[19] of a hydrogen molecule in liquid state at 14.7 K, which turns out to be ~0.85 A^2 in reasonable agreement with the experimental value[1] of 1.0 A^2. In the inset of Fig. 1(a), we have also shown the results in liquid parahydrogen. Here also the agreement is reasonably good. However, it may be mentioned that in plotting the experimental results of λ_0 $(=v\Sigma_a + D_0B^2 - CB^4$, where D_0 is the diffusion coefficient and C the diffusion cooling coefficient[16] which is related amongst other physical quantities to the second moment of neutron inelastic scattering kernel) for the parahydrogen case, use has been made only of D_0, since no value of C of parahydrogen is given.[14]

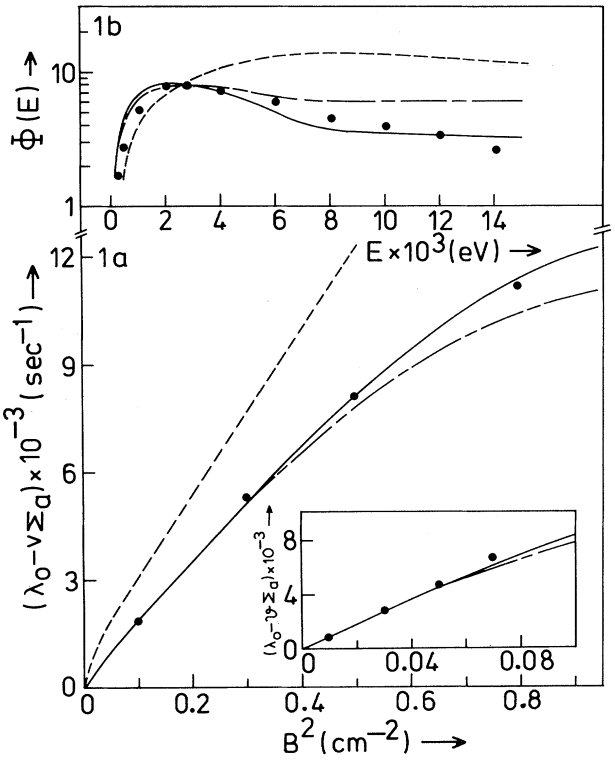

FIG. 1. (a) Comparison of calculated asymptotic decay mode for various sizes of liquid-hydrogen mixture assemblies at 20 K with the experimental results (Ref. 14). Solid circles, experimental points; dashed line, gas kernel calculations; dot dashed line, one phonon kernel; solid line, suggested kernel calculations. Inset shows the comparison for liquid parahydrogen. (b) Comparison of the calculated steady-state neutron spectra in liquid parahydrogen at 20 K with the experimental results of Whittemore (Ref. 9). Solid circles, experimental points; dashed line, gas kernel calculations; dot dashed line, one phonon kernel; solid line, suggested kernel calculations.

In order to further test our scattering kernel and critically assess the effect of chemical binding energy on neutron transport[20] we have performed the useful study of steady-state spectra,[21] $\Phi(E_i)$, of neutrons in liquid parahydrogen and para-ortho mixture in presence of $1/E$ source, $S(E_i)$. The expression for $\Phi(E_i)$ is given as[20,21]

$$\Phi(E_i) = \sum_j P_{ij}^{-1} S(E_i). \quad (2)$$

In Fig. 1(b) we have shown the calculated steady-state spectra in liquid parahydrogen at 20 K using our scattering kernel along with the experimental measurements of Whittemore.[9] The agreement between the calculated and measured spectra is quite good in the entire energy range. The effective temperature of both, the calculated neutron spectrum as well as the experimental spectrum, turns out to be ~37 K. In Fig. 1(b) also shown are the steady-state spectra calculated on the basis of one-phonon and gas kernels. The calculated spectra are very much different from the experimental spectrum and also from the one calculated using the one- plus two-phonon scattering kernel. Thus the inelastic scattering kernel incorporating only the collective motion of the liquid is able to reproduce reasonably well the detailed thermal-neutron spectrum at 20 K.

We have performed yet another study to calculate γ_{ij} which are also known to be very sensitive to the chemical binding energy incorporated in the scattering kernel.[22,23] γ_{ij} are neutron-condensed-system energy-exchange observables which are determined solely by the neutron inelastic scattering cross section.[22] γ_{ij} for liquid hydrogen at 20

K, as calculated from the suggested scattering kernel, are very much different from the ones obtained using the gas kernel, with mass of the scatterer M as 2. The value of $\gamma_{\parallel}$ which can be calculated using the experimental results of asymptotic time-dependent neutron spectra with different non $1/V$ absorbers, as has been done in the case of water,[23] obtained using the suggested kernel is equal to 0.4144 which is less than half the value (=0.9091) obtained using the gas kernel with $M=2$. If we increase the scattering mass of hydrogen molecule in gas kernel, we find that the value of $\gamma_{\parallel}$ decreases and for $M=7.5$, $\gamma_{\parallel}$ becomes nearly the same as that given by the suggested kernel. Thus the present scattering kernel with chemical binding energy may appear to be equivalent to the free gas kernel with effective mass as 7.5. However, the gas kernel with $M=7.5$ does not give the same values of other γ_{ij} as are given by the suggested kernel. Further, in the gas kernel, even with effective mass of the scatterer greater than 2, the physics of collective motion is missing.

We, therefore, conclude that the suggested kernel incorporating the chemical binding energy is able to explain various available experimental results, is also physically acceptable and is consistent with the recent experimental results of Carneiro, Nielsen, and McTague.[1] It is also clear from our study that the presence of collective motion in liquid hydrogen should be taken into account in analyzing the transport of low-energy neutrons in liquid hydrogen.[24,25]

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