Finite H$_2$ concentrations in superfluid $^4$He

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Abstract

We have studied the solubility of molecular hydrogen in bulk liquid $^4$He at zero temperature using the diffusion Monte Carlo method and realistic interatomic potentials between the different species of the mixture. Around the $^4$He equilibrium density, the H$_2$ molecules clusterize in liquid-like drops blocking the existence of a uniform dilution. On the contrary, at higher densities the cluster formation is less feasible and metastable dilute solutions may exist.

Keywords: Superfluid H$_2$; Liquid $^4$He; Diffusion Monte Carlo.

The achievement of a stable liquid phase of molecular hydrogen H$_2$ that might exhibit superfluid behaviour has been object of a continued experimental and theoretical interest. Bulk H$_2$ is a hcp solid that melts at 13.8 K whereas the critical temperature of its superfluid transition in bulk is expected to be much smaller (3-4 K) [1]. This difference has proved too large to be experimentally covered by supercooling, even in confined geometries.

We study the liquid $^4$He-H$_2$ mixture at zero temperature by means of diffusion Monte Carlo (DMC). This method solves stochastically the N-body Schrödinger equation providing results which are exact for bosonic systems like the present one. The hydrogen molecules are considered in their ground state (para-H$_2$) and interact by means of the isotropic potential proposed by Silvera and Goldman [2]. The potential of Meyer et al [3] and the HFD-B(HE) Aziz potential [4] have been used for the $^4$He-H$_2$ and $^4$He-$^4$He pair interactions, respectively. In all the calculations we have used a simulation box containing 108 particles, 5 of them being H$_2$ molecules, what suppose a macroscopic H$_2$ concentration of $\sim$ 5%.

The dilution of H$_2$ in liquid $^4$He could be favoured by its lower mass, roughly a factor two, that increases its kinetic energy with respect $^4$He. However, the well of the H$_2$-$^4$He interaction is three times deeper than the one of the He-He potential, a fact that points to a clusterization of the H$_2$ molecules in order to minimize the energy. Our results show that the latter effect dominates in a wide range of densities. The $^4$He-H$_2$ mixture has been studied at total densities 0.328, 0.365, 0.401, and 0.424 $\sigma^{-3}$ ($\sigma = 2.556$ Å), and in each case, two different types of initial configurations have been chosen. In the first type, the H$_2$ molecules are uniformly distributed in the simulation box, while in the second one, they are all close one another.

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in a cluster-like structure.

A common trend to all the densities is that the cluster structure is energetically preferred to the uniform mixture but this difference reduces when the density increases. The lifetime of the metastable homogeneous solution increases when that energy difference decreases, and therefore, a uniform mixture is more feasible at higher densities. The lifetime of the metastable homogeneous solution increases when that energy difference decreases, and therefore, a uniform mixture is more feasible at higher densities. The DMC results confirm this behaviour: the simulations starting on a uniform dilution evolve towards a molecular aggregate in the scale of a typical run. Once the cluster is formed, its structural evolution and stability depends on the density.

The two-body radial distribution function $g_{\text{H}_2-\text{H}_2}(r)$ is shown in Fig. 1 for the four densities analyzed. In all cases, the functions plotted correspond to the stable regime once the initial dilute regime has disappeared. The different behaviour between the first three densities and the highest one is reflected in the figure. For densities below 0.424 $\sigma^{-3}$ the three distribution functions are very similar, with a high central peak and a tail that approaches zero at medium distance. This structure points unambiguously to the existence of a liquid H$_2$ cluster moving in the $^4$He medium. The curve at 0.424 $\sigma^{-3}$ presents a lower central peak and values closer to one at large distances suggesting a coexistence of dilute and cluster structures.

The crossed distribution function $g_{^4\text{He}-\text{H}_2}(r)$ provides additional information on the local structure around the H$_2$ molecules. In Fig. 2, results for $g_{^4\text{He}-\text{H}_2}(r)$ at different densities are reported. In this case, the stability of the cluster structure produces a depression of the height of the first peak and a subsequent increase of the second peak that, at low densities, is even higher than the first one. On the contrary, at 0.424 $\sigma^{-3}$ one observes a behaviour much closer to the characteristic one in a homogeneous mixture.

In conclusion, only at high densities and low H$_2$ concentrations a metastable uniform 4He-H$_2$ mixture may exist. Below a characteristic density ($\sim$ 0.424 $\sigma^{-3}$) a liquid cluster of H$_2$ is predominantly observed.

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