Paramagnetic Attraction of Impurity-Helium Solids

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INTRODUCTION

Impurity-helium solids are formed when a mixture of impurity and helium gases enters a volume of superfluid helium.[1] Typical choices of impurity gas are hydrogen deuteride, deuterium, nitrogen, neon and argon, or a mixture of these. These solids consist of individual impurity atoms and molecules as well as clusters of impurity atoms and molecules covered with layers of solidified helium. The clusters have an imperfect crystalline structure and diameters ranging up to 90 angstroms, depending somewhat on the choice of impurity. Immediately following formation the clusters aggregate into loosely connected porous solids that are submerged in and completely permeated by the liquid helium. [2,3]

Im-He solids are extremely effective at stabilizing high concentrations of free radicals, which can be introduced by applying a high power RF discharge to the impurity gas mixture just before it strikes the superfluid helium. (Figure 1.) Average concentrations of 10^{19} nitrogen atoms/cm^3 and $5 \cdot 10^{18}$ deuterium atoms/cm^3 can be achieved this way. Figure 3 shows a typical sample formed from a mixture of atomic and molecular hydrogen and deuterium. Figure 6 shows typical sample formed from atomic and molecular nitrogen. Much of the stability of Im-He solids is attributed to their very large surface area to volume ratio and their permeation by superfluid helium. Heat resulting from a chance meeting and recombination of free radicals is quickly dissipated by the superfluid helium instead of thermally promoting the diffusion of other nearby free radicals. This temperature stability makes Im-He solids an ideal medium in which to study radicals that are involved

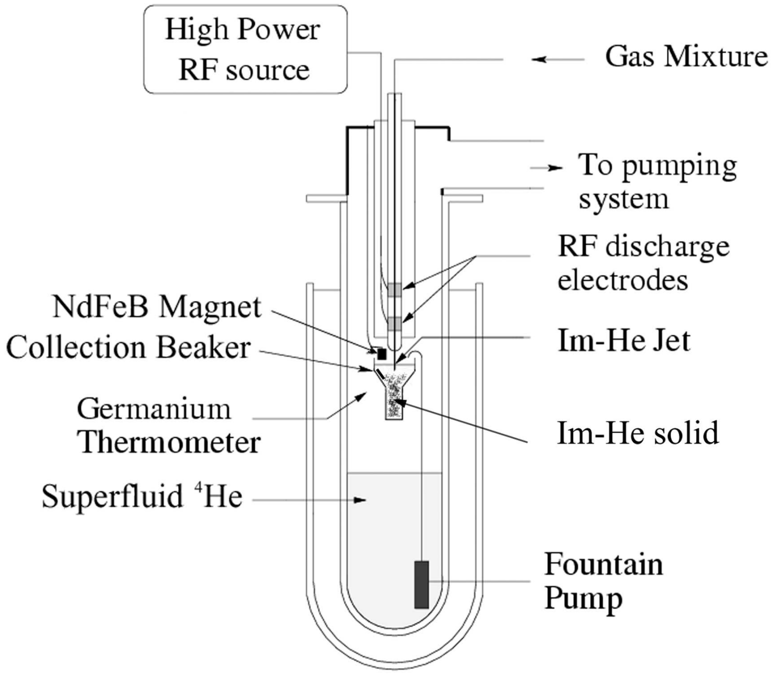

Fig. 1. Experimental apparatus for preparation and paramagnetic lifting of impurity helium solid.

in chemical reactions driven by quantum tunnelling mechanisms as opposed to conventional thermal activation.[4]

Theory of Experiment

The presence of high concentrations of stabilized radicals in Im-He solids suggests that these samples will have a substantial paramagnetic attraction to regions of high magnetic field. When a permanent magnet is lowered into and withdrawn from a beaker containing an Im-He solid sample, the submerged sample will stick to the magnet if the vertical component of the paramagnetic force exceeds the gravitational force minus the buoyant force provided by the liquid helium that surrounds and permeates the sample:

$$F_{\text{mag}} = \mu n_{\text{l}} V_{\text{c}} R(\mathbf{B}, T) \nabla \mathbf{B} \cdot \hat{\mathbf{Z}} > (\rho_{\text{c}} - \rho_{\text{He}}) V_{\text{c}} g \tag{1}$$

where F_{mag} is the vertical force exerted by the magnet on a sample, $\mu \simeq \mu_B g_e J$ is the magnetic moment of each stabilized radical, V_{c} is the total volume occupied by the impurity clusters in a fragment, n_{l} is the concentration of stabilized free radicals within volume V_{c}, $\mathbf{B}$ is the field due to the

Table 1. Characteristics of gas mixtures and impurity-helium solids.

makeup gases	respective admixture	impurities after discharge	local radical concentration, n_l
$HD{:}D_2{:}He$	1:1:100	H, D, H_2, HD, D_2	$n_l > 2.5 \cdot 10^{20}$
$D_2{:}He$	1:20	D, D_2	$2.9 \cdot 10^{18} < n_l < 2.5 \cdot 10^{20}$
$N_2{:}He$	1:100	N, N_2	$8.8 \cdot 10^{18} < n_l < 8.3 \cdot 10^{19}$
$N_2{:}He$	1:200	N, N_2	$8.8 \cdot 10^{18} < n_l < 8.3 \cdot 10^{19}$

permanent magnet, $\hat{\mathbf{Z}}$ is the vertical unit vector, ρ_c and ρ_{He} are the densities of the impurity clusters and superfluid helium, and g is the acceleration of gravity. The field and temperature dependent Brillouin function[5]

$$R(\mathbf{B},T) = \left(1+\frac{1}{2J}\right)\coth\left[\left(J+\frac{1}{2}\right)\left(\frac{\mu\mathbf{B}\cdot\hat{\mathbf{Z}}}{JkT}\right)\right] - \frac{1}{2J}\coth\left[\frac{\mu\mathbf{B}\cdot\hat{\mathbf{Z}}}{2JkT}\right] \quad (2)$$

accounts for the thermalization of the magnetic moments in the sample, where J is the angular momentum quantum number of the radicals, k is Boltzmann's constant and T is the temperature. Note that $M_z = \mu n_l R(\mathbf{B},T)$ and $M_z = \chi\mathbf{B}\cdot\hat{\mathbf{Z}}$ when $\mu\mathbf{B}\cdot\hat{\mathbf{Z}}/kT \ll 1$.

Calculating the force needed to lift samples above the surface of the superfluid helium is complicated by the fact that surface tension causes retention of a substantial amount of liquid helium within the pores of the sample. If the weight and volume of the small amount of helium solidified on the surface of the clusters is ignored, the force needed to withdraw a sample from the helium is:

$$F_{\text{mag}} = \mu n_l V_c R(\mathbf{B},T)\nabla\mathbf{B}\cdot\hat{\mathbf{Z}} > (\rho_c + \frac{V_p}{V_c} f\rho_{He})V_c g \quad (3)$$

where V_p is the volume of the pores in the sample fragment and $f = 0.23$ is the degree of saturation of those pores. The porosity V_p/V_c varies greatly from sample to sample, but X-ray studies suggest that $V_p/V_c = 150$ is typical of deuterium samples and $V_p/V_c = 220$ is typical of nitrogen samples.[2,3] Measurements of the volume of liquid helium displaced by impurity-helium solids suggest an estimate of $f = 0.23$. [6]

RESULTS

We formed samples at $T = 1.5$ K using the four gas mixtures listed in Table 1. Deuterium hydride sample formation and nitrogen sample formation are shown in figures 2 and 5 respectively. Submerged samples within two to three millimeters of the magnet clung to it tightly in each of the experiments. Fragments of submerged deuterium-helium solid and nitrogen-helium solid are shown clinging to the magnet in figures 4 and 7 respectively. At this distance our magnetic field gradient is about 1 T/cm and $\mathbf{B} \sim 1/2$ T. However only the smallest fragments of the HD:D_2:He sample stayed in contact with the magnet as it was lifted above the helium surface. If we make the reasonable assumption that our cluster densities are comparable to those of corresponding bulk frozen impurity gases, we find the ranges for n_l listed in Table 1.

CONCLUSION

The experiments show that modest magnetic field gradients (∼1 T/cm) can exert substantial forces on impurity helium solids. These techniques may be applied in future experiments to move Im-He solids from place to place or to sort them by the concentrations of radicals within their clusters. Refinements of this experiment using magnets specifically shaped to have constant field gradients could greatly improve our crude estimates of n_l. To date the formation of pure hydrogen Im-He solids has been hindered by the fact that solid hydrogen floats in superfluid helium. Our group will attempt to form a hydrogen Im-He solid using a magnetic field gradient to counteract the buoyant force and pull the sample into the liquid helium as it is formed.

ACKNOWLEDGMENTS

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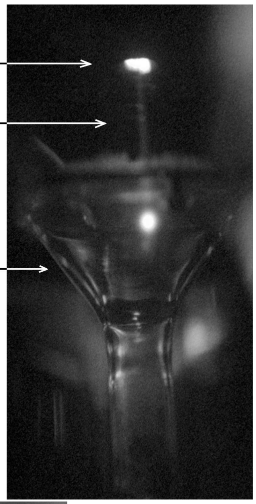


Figure 2.

Right: Formation of sample with initial gas mixture HD:D_2 :He = 1:1:100, producing H-D-HD-D_2 -He sample.

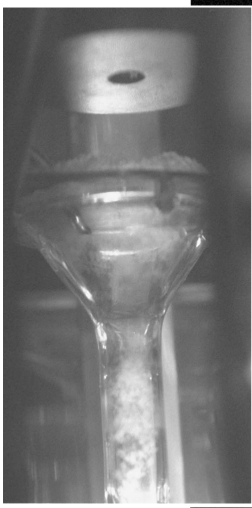


Figure 3.

Left: Stabilized H-D-HD-D_2 -He sample submerged in superfluid helium following formation.

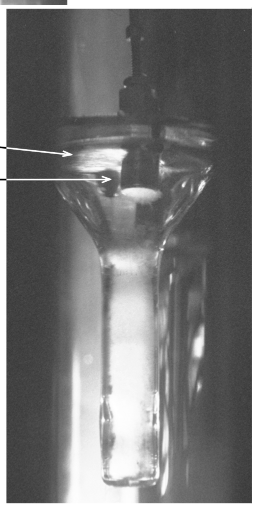
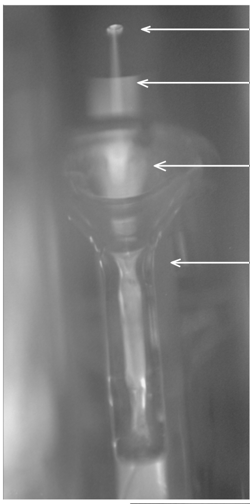


Figure 4.

Right: A D-D_2 -He sample formed from initial gas mixture D_2 :He = 1:20 using RF discharge clings to the magnet.

Figure 5.

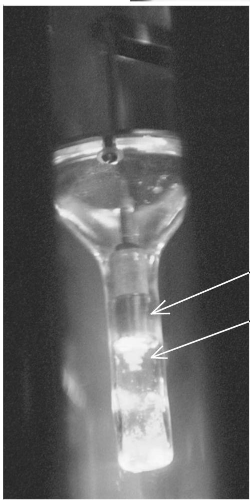


Left: Formation of N-N_2 -He sample using initial gas mixture N_2 :He = 1:200 and RF discharge.

Figure 6.

Right: Stabilized N-N_2 -He sample submerged in superfluid helium following formation.

Figure 7.



Left: An N-N_2 -He sample formed from an initial gas mixture N_2 :He = 1:200 using RF discharge is lifted by the magnet.