baryons or mesons are produced are likely to exhibit bound B_c nuclei.

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Finally one notes the strangeness analog states discussed by Kerman and Lipkin⁸ will have their counterparts in charm analog states, the latter resulting from the extension of the Sakata model to SU(4).

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Hydrogen Surface Contamination and the Storage of Ultracold Neutrons

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> The results of hydrogen profile measurements on samples prepared by the same methods used for these materials when making ultracold-neutron (UCN) storage bottles show that the surface hydrogen is sufficient to account for the bulk of the anomalous shortening of UCN storage times.

Ultracold neutrons (UCN) are neutrons with such low energy (~ 10^{-7} eV) that they undergo total reflection for all angles of incidence from many materials. As such, these neutrons can be stored in "bottles" for substantial periods of time and may provide a source of neutrons upon which a variety of important fundamental measurements can be made, e.g., the accurate determination of the neutron lifetime¹ and a more sensitive search for its electric dipole moment.²

The first direct measurements of ultracoldneutron storage times³ revealed serious discrepancies between the observed storage times, τ_s , and those expected for an ideally flat, pure surface. While neutrons below a critical energy undergo total reflection, there are mechanisms which result in the "loss" of these neutrons. During a reflection, the neutron can be lost via nuclear capture or by being inelastically scattered to an energy above the critical energy.

Further experiments⁴⁻⁸ have shown similar

anomalies and much effort has been devoted to understanding of these results.^{6,7,9-11} Several authors^{3,6,7,12} have concluded that since the observed storage times appear to be independent of temperature, surface impurities cannot play an important role in the UCN losses. The only published data on this point, Fig. 1 (Refs. 7a and 7c), is indeed consistent with UCN losses being independent of temperature, but it does not rule out variations in $\tau_{\rm c}$ up to a factor of 2 over the temperature range covered (dotted line, Fig. 1).

It is difficult to predict a unique temperature dependence of τ_s if impurities are responsible for the anomalies. If the impurity concentration was temperature independent and if the losses mainly result from inelastic scattering from a coherent scatterer or harmonically bound nucleus. the loss rate will be proportional to the Planck oscillator occupation number, i.e., $\sim T$ for large T and $\sim \exp[-\omega_0/T]$ for small T (with respect to the oscillator frequency ω_0).¹³ For incoherent



FIG. 1. The number of UCN remaining in a glass bottle vs storage time with various bottle-wall temperatures: room temperature (\bullet), temperature varying along the length of the storage bottle between 200°C and 500°C (\bigcirc), and temperature varying along the length of the storage bottle between -40°C and -170°C (×). The dashed line corresponds to twice the storage time of the solid curve. Data are from Refs. 7a and 7c.

scattering the behavior depends on the phonon density of states; a Debye spectrum gives a loss ~ $T^{7/2}$ for $T \leq \Theta_D$ where Θ_D is the Debye temperature. Common pure materials show measured total-cross-section variations¹⁴ that vary as T^n with *n* between 0.1 and 2 for $100 \leq T \leq 1000^{\circ}$ K. The variation of impurity concentration with *T* may be expected to have the opposite effect on loss rates.¹⁵ Given these complications, a surface model with a behavior consistent with Fig. 1 is not very hard to produce.

Hydrogen is the prime candidate for causing the UCN loss because of its abundance and its large neutron scattering cross section, but chlorine has also been mentioned as a possibility.^{10,11} We have measured the hydrogen concentration versus depth for various samples which we prepared by the same methods used for the preparation of UCN experiments. Copper samples were electropolished in H_3PO_4 as in Ref. 9. The highalumina, boron-free glass was manufactured at the Hirst Research Center (GEC) to the formula of Soviet 13-B glass and then polished and repeatedly etched in 3% HF.^{6,16} The graphite sample, supplied by A. Steyerl, had been manufactured, outgassed, and handled in the same way as the graphite used in his UCN experiments.⁴

The hydrogen surface contaminations were measured by bombarding the samples with ^{15}N and measuring the yield of 4.43-MeV γ rays from

the ${}^{15}N + H \rightarrow {}^{12}C + {}^{4}He + 4.43$ -MeV γ -ray resonant reaction.¹⁷ Because there is a narrow, isolated resonance with a large peak cross section in this reaction, the profile of the hydrogen concentration versus depth can be determined by measuring the yield of 4.43-MeV γ rays versus energy of the ¹⁵N. This technique has been used to quantitatively analyze the hydrogen content of a number of materials¹⁸ and the accuracy of the method has been confirmed by comparison with other methods.¹⁹ Perhaps it should be noted that when measuring the total amount of H/cm^2 on a surface, the calibration of such a method is independent of the material being analyzed. Because of the finite width of the resonance, this method has a finite depth resolution (σ_r). The depth resolutions for copper, graphite, and glass are 30, 52, and 66 Å, respectively. The UCN penetrates into these solids by a distance of order ~100 Å comparable to the depth resolution of the method.

The measured hydrogen profiles for copper, graphite, and high-alumina glass are shown in Fig. 2. These figures show the profiles as measured with no corrections for the depth resolution of the method. The concentration scale had been previously determined by using a calibration sample; these results should be accurate to better than 10%. All these samples were profiled both at room temperature and after heating the sample for various lengths of time at different temperatures from 190-265°C. This heating was carried out in the target chamber, and the hydrogen profiles were then measured without removing the sample. The profiling required that the heating element be disconnected, but the samples were periodically reheated; hence, the profiles were measured at nearly the same temperature as the initial heating. The chamber was pumped with liquid-nitrogen-trapped ion pumps to a vacuum of about 7×10^{-7} Torr.

In addition to the data shown in Fig. 2 for the copper samples, room-temperature profiles were measured following a variety of surface treatments including washing, mechanical polishing and electropolishing, and all combinations of these. These profiles were qualitatively similar to that shown for copper at room temperature in Fig. 2 but with the peak concentrations varying from about 0.7 to 1.3×10^{23} atoms/cm³. While there was no simple correlation between treatment and hydrogen content, mechanical polishing appeared to increase the hydrogen near the surface.

We had considerable difficulty measuring the



FIG. 2. The hydrogen profiles for copper (mechanically polished, electropolished and washed in acetone and alcohol), pyrolytic graphite [baked at 400°C for one week in a vacuum of 10^{-6} Torr and then exposed to air for about 6 months (Ref. 4)] and glass (Refs. 6 and 16) (etched in HF). After the copper and graphite samples were profiled at room temperature (×), they were reprofiled without exposure to air after heating to 190°C for 18 hours (\bigcirc) and after heating to 265°C for an additional 13 hours (\bigcirc). The glass was profiled after heating to 210°C for 2.5 days. The dashed curve represents the *minimum* hydrogen content consistent with the data (see text).

hydrogen profile for the glass sample. This glass is an excellent insulator and, as a result, the ¹⁵N beam electrostatically charged the surface, changing the ¹⁵N energy as it approached the surface. We did not put any conductive coating on the glass because to do so might introduce considerable hydrogen. As a result, repeat measurements at the same ¹⁵N energy did not give the same result. The bars shown on Fig. 2 for glass represent this spread. This electrical charging makes interpretation of the results for glass more difficult than those for copper or graphite. The most important question is what is the peak concentration. To get a lower limit on this concentration we lowered the beam intensity to 0.2nA and tried to keep the beam constant. Under these conditions, we obtained a count rate corresponding to a peak concentration of $C_0 = 3.2 \times 10^{22}$ H atoms/ cm^3 with good reproducibility. This peak count rate must correspond to a total hydrogen content of at least that of an infinitely thin hydrogen layer which gives this count rate. For this case one can integrate the Breit-Wigner

cross section to get a minimum for the total hydrogen content of $\frac{1}{2}\pi C_0 \sigma_x = 3.3 \times 10^{16}$ H atoms/cm². The dashed curve in Fig. 1 for glass corresponds to the expected profile for this *minimum-concentration case*, normalized to the peak concentration of 3.2×10^{21} cm⁻³.

Using these data we estimate the average UCN loss probability per bounce, $\overline{\mu}$, from these surfaces and compare the results with the UCN meas urements. Ignoring the losses, we calculate the unperturbed wave function in the one-dimensional potential^{6,7,12} $V(z) = K \sum_i N_i(z) a_i$, where the sum is over all constituents of the material, including impurities, $N_i(z)$ is the number density, and a_i the coherent scattering length of the *i*th species. Then we calculate μ from

$$\mu = \sum_{i} \sigma_{L}^{(i)} \int N_{i}(z) |\psi(z)|^{2} dz , \qquad (1)$$

where $\sigma_L^{(i)}$ is the total loss cross section for UCN. Averaging μ over angle and energy, we obtain $\overline{\mu}_{\circ}$ We assume the materials consist of two regions-the bulk and a surface layer with a depth derived from the hydrogen profile data-with each region homogeneous and sharp boundaries defining all regions. The situation is complicated by our lack of knowledge of the form of the surface H. This means both that $\sigma_L^{(H)}$ in (1) is uncertain because it depends on the binding of the H. and that the densities necessary to calculate V(z) depend on whether the H is, e.g., in some hydrocarbon [which may have a negative V(z), which will result in a large UCN loss] or, e.g., distributed interstitially in the bulk lattice. We will make the most conservative assumption in the cases of copper and glass, namely, that the H is simply inserted into the bulk material without changing its density. We take $\sigma_L^{(H)}$ for the H as 50 b at a neutron wavelength of 10 Å and, in the case of copper, a H density of 1×10^{23} cm⁻³ over a thickness of 25 Å. This is about half of the total surface H at room temperature but equal to that observed at $200^{\circ}C$ [Fig. 2(a)]. We thus calculate $\mu = 5.2 \times 10^{-4}$ as compared to measured values of 5.9×10^{-4} (Refs. 7 and 18) and $(5-8) \times 10^{-4}$ (Ref. 16). The calculated velocity dependence of μ agrees quite well with the data of Groshev *et* al_{\circ}^{20} For glass, we take a H density of 3×10^{22} cm⁻³ with a thickness of 50 Å, which gives about half the minimum surface H compatible with Fig. 2(c). The calculation gives $\vec{\mu} = 1.6 \times 10^{-4}$ compared to measured values of 2×10^{-4} (Ref. 6) and 1.57×10^{-4} (Ref. 16). For the graphite, we assume a film of typical hydrocarbon (enabling the

H to have the maximum effect on UCN loss) with a H density of 7×10^{22} cm⁻³ and a thickness of 30 Å, producing a total coverage of 2×10^{16} cm⁻², which is about what we get from the surface room-temperature peak of Fig. 2(b). We take a bulk H density of 1×10^{22} cm⁻³ and calculate μ = 2.9×10^{-4} compared to the measured⁴ 8×10^{-4} . Pure graphite would give $\mu = 6 \times 10^{-6}$, theoretically; so the H accounts for some but not all of the anomaly. We should stress that the vacuum conditions in our measurements (7×10^{-7} Torr) were certainly no worse than those in the UCN storage experiments; 10^{-4} Torr in Ref. 4 and 16.

Several authors^{6,7,12} have emphasized the similarity of the observed $\overline{\mu}$ for surfaces with markedly different theoretical expectations, i.e., to within a factor of ~ 10 all surfaces look the same to UCN. Similarly, based on our data and the work of others,¹⁵ who find similar or greater amounts of surface hydrogen in various materials, one is tempted to say that again, to within a factor of about 10, all surfaces look the same to hydrogen profiling measurements.

Although the only way to *prove* that impurities are responsible for the UCN losses is by UCN experiments (perhaps at cryogenic temperatures and using materials such as solid oxygen¹¹), the present work certainly shows that, contrary to the conclusion of some authors,^{3,6, 7,12} hydrogen contamination can account for the bulk of the observed anomalies and presents serious problems for the storage of UCN.

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