γ-ray spectrometry study of the heating of ultracold neutrons as they interact with a surface

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A direct experiment was carried out to determine the reasons for the anomalous heating of ultracold neutrons as they interact with the surface of a material. The hypothesis that hydrogen is the main cause of this heating has been confirmed by a γ-ray spectrometry method.

As ultracold neutrons interact with the surface of a material, they tunnel into the material to an effective depth on the order of 100–300 Å. As a result, some of them are absorbed by nuclei of the material or undergo inelastic scattering (a heating) and leave the region of ultracold energies. If there are impurities at the surface, the probabilities for these processes may be substantially higher than that calculated for a "clean" surface. Numerous experiments on the storage of ultracold neutrons in vessels of various materials have shown that it is not possible to achieve the theoretical probability for the capture and heating of ultracold neutrons. To a large extent, this effect has stemmed from an additional or "anomalous" heating of the neutrons.[1] In an effort to explain this heating, it has been hypothesized that there is a universal hydrogen-containing surface layer for all materials (Ref. 2, for example). So far, however, no direct experiment with ultracold neutrons has been carried out which would unambiguously establish that there is a relationship between the probability for an additional heating of the neutrons in collisions with the surface, on the one hand, and the hydrogen concentration at this surface, on the other.

In an attempt to solve this problem we have carried out experiments to detect a γ-ray emission which would arise during the interaction of ultracold neutrons with the surface of stainless steel, which is the material used most commonly for vessels and neutron ducts for ultracold neutrons. Our basic goals were to determine the hydrogen concentration, from the yield of 2.23-MeV γ rays from the reaction $n(p, d)\gamma$, and to establish a correlation between this quantity and the probability for inelastic scattering of ultracold neutrons.

Figure 1 shows the experimental layout. The apparatus (Fig. 1a) consists of a vacuum chamber with the test sample, a γ-ray spectrometer, and a detector of ultracold neutrons. Neutrons enter the chamber from a neutron transport duct of the ultracold-neutron apparatus. This apparatus provides an output flux density of 3–4 $n/(\text{cm}^2 \cdot \text{s})$ in the velocity interval 3.2–0.6 m/s. The ^{3}He-based ultracold-neutron detector, with an entrance window 60 cm^2 in area, is connected to the chamber through a diaphragm with an aperture with an area $S_0 = 1$ cm^2. The test sample is a stack of plates of type 12Kh18N10T stainless steel, 60×100 mm^2 in area and 100 μm thick. The total area of the plates is $S_s = 0.9$ m^2; the thickness of the gap between plates

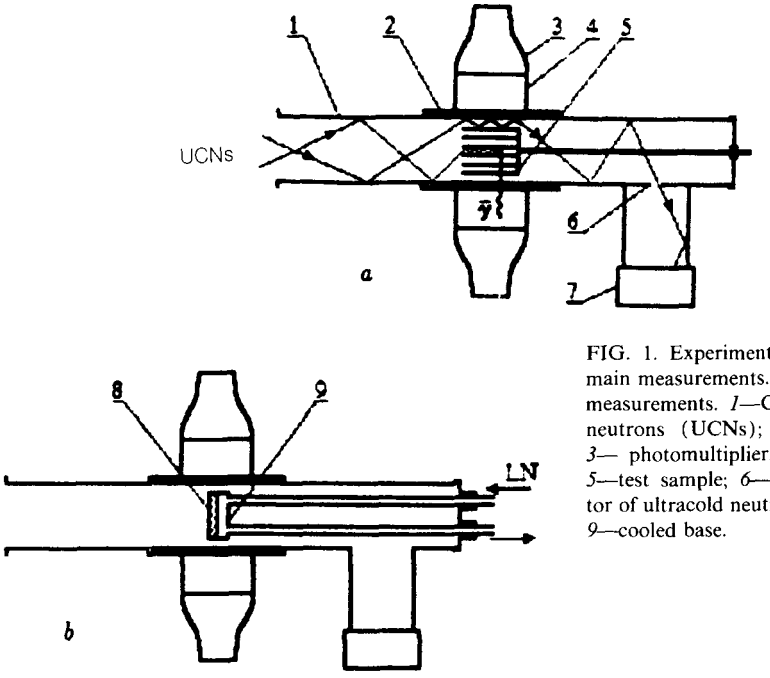

FIG. 1. Experimental layout. a: For the main measurements. b: For the calibration measurements. *1*—Chamber for ultracold neutrons (UCNs); *2*—converter screen; *3*— photomultiplier; *4*—NaI(Tl) crystal; *5*—test sample; *6*—diaphragm; *7*—detector of ultracold neutrons; *8*—polyethylene; *9*—cooled base.

is 1.5 mm. The γ-ray spectrometer is made from two scintillation blocks based on NaI(Tl) crystals 75×80 mm in size. Between the walls of the chamber and the crystals is a special converter screen: a flat layer of boron carbide 1 cm thick. This converter screen, combined with the γ-ray spectrometer, constitutes the detector of the neutrons which are heated in the interaction with the surface of the test sample. As the heated neutrons strike the converter, they are captured, and 477-keV γ rays from the reaction $n+{}^{10}\mathrm{B}\rightarrow{}^{7}\mathrm{Li}+\alpha+\gamma$ (477 keV) are detected by the γ-ray spectrometer. The entire apparatus is surrounded by a lead shield and a boron-loaded polyethylene shield.

When the test sample is exposed to the ultracold neutrons, peaks representing the total absorption of γ rays with energies of 477 keV and 2.23 MeV appear in the γ-ray spectrum. The probability that a neutron will be heated when it collides with the surface, μ_{ie}, and the probability that it will be captured by a proton, μ_c, are given by

$$\mu_{ie}=J_{ie}/\epsilon_{ie}S_s\Phi=J_{ie}\epsilon_n S_0/J_n\epsilon_{ie}S_s, \tag{1}$$

$$\mu_c=J_c/\epsilon_c S_s\Phi=J_c\epsilon_n S_0/J_n\epsilon_c S_s. \tag{2}$$

Here $\Phi = J_n/\epsilon_n S_0$ is the flux density of ultracold neutrons bombarding the sample, J_n is the count rate of ultracold neutrons at the detector, ϵ_n is the efficiency at which neutrons which have passed through the hole in the diaphragm are detected, ϵ_{ie} is the efficiency at which the neutrons heated at the surface of the test sample are detected on the basis of the count in the peak representing the total absorption of 477-keV γ rays, and ϵ_c is the efficiency at which 2.23-MeV γ rays are detected. In order to

determine the absolute values of μ_{ie} and μ_c, we need to know the efficiency ratios ϵ_n/ϵ_{ie} and ϵ_n/ϵ_c. These ratios were measured with the help of a calibration sample, a polyethylene disk 3 mm thick and with an area S_p. This disk was put in place of the test sample (Fig. 1b); it was mounted in the chamber on a stainless-steel base whose temperature could be varied over the interval 80–300 K. Between the dish and chamber walls was an annular gap with an area $S_k=2$ cm^2, through which some of the ultracold neutrons reached the detector. In the calibration measurements, the diaphragm was removed, and the efficiency at which the ultracold neutrons which passed across the annular gap were detected was ϵ_n.

The neutrons incident on the polyethylene were completely heated or captured. The relative contributions of these processes are found as σ_{ie}/σ_t and σ_c/σ_t, where σ_c, σ_{ie}, and σ_t are the capture cross section, the inelastic cross section, and the total cross section, respectively. By measuring the count rates J_{ie} and J_c in the peaks representing the total absorption of γ rays with 477 keV and 2.23 MeV, we find the ratios we need:

$$\epsilon_{ie}/\epsilon_n=\sigma_{ie}S_kJ_{ie}/\sigma_tS_pJ_n, \tag{3}$$

$$\epsilon_c/\epsilon_n=\sigma_cS_kJ_c/\sigma_tS_pJ_n. \tag{4}$$

The only quantity here which has not been determined is σ_{ie} (the value of σ_c for the CH_2 molecule is 290 b for an average neutron velocity of 4.7 m/s). To measure σ_{ie}, we cooled the disk to about 80 K and then determined the changes in the count rates, $\Delta J_{ie}$and ΔJ_c, due to the decrease in the cross section σ_{ie} as the temperature was lowered from 300 to 80 K. Since we have $\Delta J_{ie}/\Delta J_c = \epsilon_{ie}/\epsilon_c$, by using (3) and (4) and the measured γ-ray spectra for polyethylene we can determine σ_{ie}, ϵ_{ie}/ϵ_n, and ϵ_c/ϵ_n. From the data of our measurements we find $\sigma_{ie}=3745\pm370$ b at room temperature, and we find the ratios $\epsilon_{ie}/\epsilon_n=(9.2\pm0.06)\%$ and $\epsilon_c/\epsilon_n=(2.00\pm0.15)\%$.

The γ-ray spectra were measured in various stages of a multistep removal of impurities from the surface of the test sample. In the first step, the sample was washed with acetone. In the second, it was electropolished in H_3PO_4 and then held in the atmosphere for 1.5 months. In the third step, it was electropolished again. In the fourth step it was annealed in vacuum at 350°C for 6 h. The heights of the peaks at 477 keV and 2.23 MeV turned out to be very sensitive to the degree to which the surface of the test sample was cleaned. Figure 2 shows fragments of γ-ray spectra near these peaks in all four steps of the surface cleaning. As the cleaning proceeds, the probability for the heating of ultracold neutrons is seen to decrease; at the same time, there is a decrease in the hydrogen concentration at the surface. After the annealing, the hydrogen concentration decreases to the extent that the peak representing the total absorption of 2.23-MeV γ rays can no longer be seen at all, although the peak representing the 477-keV γ rays is still present. The small (20%) addition to this peak is due to 512-keV annihilation γ-rays. Analysis of the spectra by means of expressions (1) and (2) shows that μ_e and μ_c are related in a linear fashion (Fig. 3). This result is unambiguous evidence that hydrogen on the surface of the steel is the primary reason for the additional inelastic scattering of the ultracold neutrons. On the other hand, this heating mechanism is not the only one operating, since even when the hydrogen concentration is zero ($\mu_c=0$), the quantity μ_{ie} remains nonzero, at (0.95

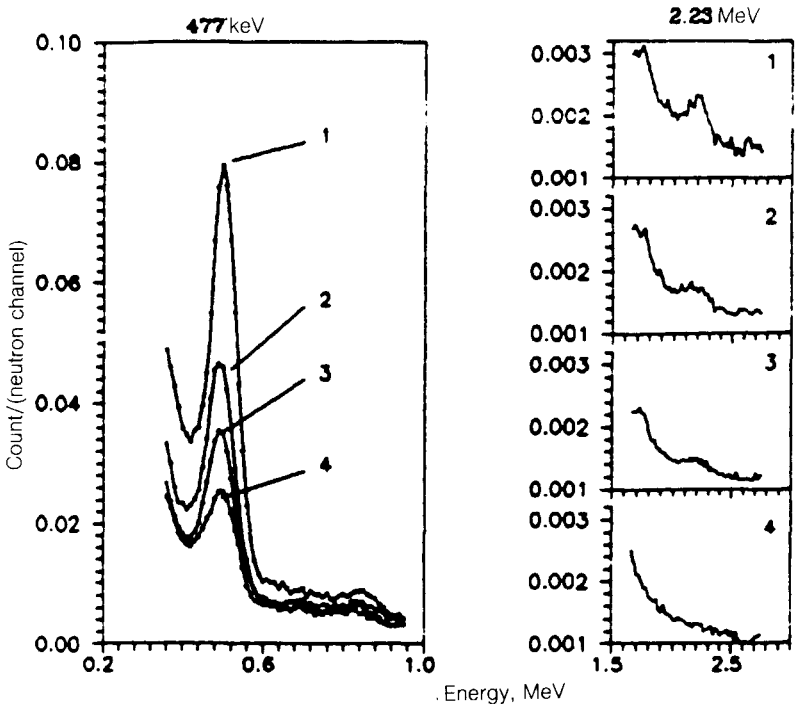

FIG. 2. Fragments of the γ-ray spectra near energies (E) of 477 keV and 2.23 MeV at various stages of the cleaning of the surface of the test sample. *1*—After the washing with acetone; *2*—after the first electropolishing; *3*—after the second electropolishing; *4*—after annealing in vacuum.

$\pm 0.06) \times 10^{-4}$. The calculated value of μ_t (the total loss coefficient) for stainless steel is 1.44×10^{-4} at an average velocity $\bar{V} = 4.7$ m/s and is determined by capture. The nature of the residual "hydrogen-free" heating thus requires further research. We do not rule out the possibility that this process is a heating of the ultracold neutrons on the stainless steel which is intensified for some reason, or that it is a heating by surface impurities which do not contain hydrogen and which do not desorb during annealing.

For a quantitative estimate of the surface concentration of hydrogen we used the expression given in Ref. 2:

$$\mu_c = 8 N_H \bar{V} V_{th} \sigma_{th} / 3 V_{lim}^2, \tag{5}$$

where N_H is the number of hydrogen atoms per unit area, V_{th} is the velocity of thermal neutrons, σ_{th} is the cross section for the capture of thermal neutrons by hydrogen, and V_{lim} is the limiting velocity for stainless steel. It can be seen from Fig. 3 that the surface concentration of hydrogen is reduced by the cleaning from 1.6×10^{17} atoms/cm^2 to $N_H < 6 \times 10^{15}$ atoms/cm^2. This result is consistent with data found by a nuclear-reaction method.[3]

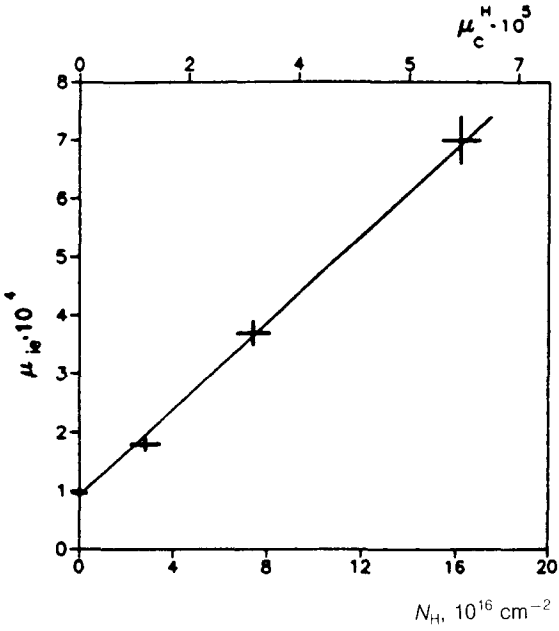

FIG. 3. Probability for the heating of ultracold neutrons (μ_{ie}) versus the probability for the capture of a neutron by a proton (μ_c) and versus the hydrogen concentration on the surface (N_H).

The method proposed here can be used effectively to study the interaction of ultracold neutrons with other materials, in particular, to study the elevated loss of ultracold neutrons observed[4,5] in the case of low-temperature beryllium vessels. In general, this study has demonstrated that it is possible to develop a new direction for studying surfaces: neutron-radiation analysis of the nuclear composition of the surface with the help of ultracold neutrons. The use of intense sources of ultracold neutrons, combined with Ge(Li) detectors, should greatly expand the range of problems which can be solved by neutron-radiation and possibly neutron-activation analysis of surfaces with the help of ultracold neutrons.

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