Model of the gamma ray-induced out-gassing in the nn-scattering experiment at YAGUAR

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Abstract

A first attempt to measure the neutron-neutron scattering length directly has been performed at the pulsed reactor YAGUAR [1]. An unexpectedly large background that changed quadratically with the reactor power (the very feature that distinguishes n-n scattering) was observed. Detailed shape analysis of the time-of-flight spectra for the nn- and n^{3}He-runs gave evidence of radiation-induced out-gassing of molecular hydrogen and possibly water from the walls of the measuring chamber during the reactor pulse [2]. Here we propose a model for a gas desorption induced by a low-energy electrons which are created by gamma radiation in the aluminum wall of the nn- chamber during the reactor pulse. The model agrees with other out-gassing data and can help to find ways of mitigating the gas desorption effect in the direct n-n scattering measurement.

Introduction

The DIa_{nn}A (Direct Investigation of a_{nn} Association) is using the unique pulsed, high-intensity YAGUAR reactor to make the first direct measurement of the $^1S_0$ neutron-neutron scattering length a_{nn}. Indirect measurements of a_{nn} are challenging and have yet to agree within experimental uncertainty [3, 4, 5]. The development and progression of the DIa_{nn}A experiment is well-documented [6, 7, 8] with specifics of the experiment often presented at ISINN conferences [9, 10, 11, 12, 13, 14, 15, 16, 2].

The experiment uses the time-of-flight (TOF) method with a 12-m flight path to separate thermal from fast neutrons which is well collimated to reduce multiply scattered thermal neutrons. The thermal neutrons interact within an evacuated through-channel that runs axially through the center of the annular YAGUAR reactor. The reactor uses a 90% enriched ^{235}U salt water solution as the neutron source with an average energy per pulse of 30 MJ over a pulse duration of 900 μs [17]. This translates to a neutron density of 10^{13} per cm^3. A fraction of the nn-scattered thermal neutrons reach a well-shielded neutron detector at the end of the TOF beamline.

The detector counts per pulse, N_{nn}, integrated over the thermal part of the TOF spectrum, is given by [14]:

$$N_{nn} = (0.96 \pm 0.02)\pi\, a_{nn}^2 \frac{F^2}{\tau\, v_{th}}\, V\, \Omega_{\text{eff}} \ \text{(counts/pulse)}, \qquad (1)$$

where τ is the FWHM duration of the reactor pulse, v_{th} is the *average* velocity of the Maxwellian spectrum, $F = \int \Phi(t)dt$ is the neutron fluence of the total pulse, V is the nn cavity volume, and $\Omega_{\rm eff}$ is the dimensionless value of the detector solid angle. The value (0.96 ± 0.02) was calculated numerically in [14] and represents a weakly-varying function that accounts for space-temporal variations of the neutron flux given a realistic shape of the YAGUAR 900 μs pulse at a maximal power. With the parameter values for our experiment we expect approximately $N_{nn} \simeq 170$ nn counts per one burst. However, preliminary neutron-neutron scattering measurements [1] show a count rate per pulse that is 30 times higher as shown in Figure 1. This large thermal background signal has an approximately quadratic dependence on the reactor pulse energy as expected for the nn-scattering signal.

Figure 1: The data points represent time-of-flight data for neutron-neutron scattering at a reactor pulse energy of 31 MJ and a 12-m flight path. The overall count rate is approximately 30 times higher than predicted. The solid line is a fit to the data with a Maxwellian neutron speed distribution, a constant n-n cross section and appropriate detector efficiency.

An anomalously high detector count and the poor fit to the experimental data as shown in Figure 1 can be a result of a pressure increase in the nn measuring chamber during the YAGUAR pulse due to desorbed molecules of H_2 and H_2O that are liberated from the aluminum wall separating the chamber and the neutron moderator. In such a case, the detector responce depends quadratically on the reactor power (which we can vary) as demonstrated in [2]. Any other source of background, thermal neutrons scattering from collimators or beamline, for example, varies linearly with pulse energy. Besides, both H_2 and vapor H_2O have scattering cross sections that are nearly $1/v$ dependent in the thermal energy region, as demonstrated in [18]. Figure 2 shows a much improved fit to the experimental data assuming the desorbed gas is H_2. A similar fit assuming H_2O is given in [2].

Following Dobrozemsky [19] who measured a desorption rate for unbaked Al of $q_r = 4.5 \times 10^{-11}$ torr-l/(cm^{3}s MR/hr), we estimate that the 2.6×10^6-Rad gamma radiation in YAGUAR reactor pulse causes the molecular hydrogen desorbed density $n_{H_2} \sim 3 \times 10^{13}$ molecules/cm^3. We can compare such predicted desorbed gas density to our actual data. Assuming the desorbed gas is exclusively H_2, we can express the count rate at the detector N_{det}, which is about 4200 counts per pulse, in terms of the density of H_2,

$$N_{det} = \sigma_{H_2}\, n_{H_2}\, F\, V\, \Omega_{\rm eff} \quad \text{(counts/pulse)}, \tag{2}$$

where $F = 1 \times 10^{15}$ n/cm^2 is the neutron fluence per pulse, V is the nn cavity volume of 1130 cm^3, $\sigma_{H_2} = 70$ b [18] is the thermal neutron cross section per hydrogen molecule

Figure 2: Neutron-neutron scattering data assuming desorption scattering from H_2 gas. The data points are identical to those in Figure 1 but the solid line fit is to a Maxwellian distribution, the appropriate detector efficiency and the velocity dependent scattering cross section of H_2.

and $\Omega_{\rm eff}$ is the dimensionless value of the detector solid angle (5×10^{-6}) [6]. This gives a desorbed gas density of $n_{H_2} \sim 1.4 \times 10^{13}$ molecules/cm^3 which is not far away from the above estimate having in mind possible different conditions for gas adsorbing on walls.

Radiation induced desorption

Desorption from metal surfaces by low-energy (15−200 eV) electrons was discovered by Menzel and Gomer [20] and, independently, by Redhead [21] in 1964. The study of the photon and synchrotron radiation-induced desorption soon followed. Though short-lived, dynamic pressure rises in accelerator beamlines were first noticed as early as 1973 at CERN, yet even by 1997 the phenomenon "was not understood" [22]. The effect, measured at accelerator facilities worldwide, is now characterized by the desorption yield η, with units of desorbed molecules (or other gas constituents) per impact particle. The radiation induced desorption, or "non-thermal outgassing", falls into three categories: photo-induced, electron-induced, and ion-induced. Workshops solely devoted to the specific case of ion-induced (beam) desorption at accelerator facilities began in 2003 in order to keep up with the technical demands for designing facilities like the Large Hadron Collider [23].

As suggested in [2], the H_2 desorption in the YAGUAR *nn* experiment is probably due to the low-energy electrons which are created by gamma radiation in the aluminum wall of the nn- chamber during the reactor pulse. The goal of this report is not to examine the mechanisms through which the low energy electrons cause desorption (possible mechanisms are still debated by experts), but rather to describe a model for estimating η_γ which is effective in the n-n experiment.

In general, the desorption yield η spans many orders of magnitude and depends on both the properties of the incident particle (type, energy, angle of incidence, fluence) as well as the properties of the surface (composition, treatment techniques such as baking, etching, coatings). One recent and comprehensive review of recent progress in this field is given by Hilleret [24]. It is now realized that the worst material with repect to radiation-induced outgassing is alumnimum. In comparison with stainless steel, which typically has η-values 5−10 times less. For baked stainless steel low energy electrons produce typical values of $\eta_e \simeq 0.2$, while 20-keV x-ray photons lead to $\eta_{\rm x} \simeq 0.01$. The smaller value for these photons is due to the fact that only part of photon energy is converted to electrons in the material. The desorption coefficient for heavy ions of hundreds keV energy (for example $^{39}K^+$) was found to be large, $\eta_{\rm K}$ (∼10000), and scale with the **electronic** energy loss dE/dx of the heavy ion. Huge amount of produced "soft" (∼100-eV) δ-electrons per one $^{39}K^+$) ion is responsible for such large $\eta_{\rm K}$ value.

Recently Molvik et al. [25], in the study of molecular desorption from stainless steel performed with beams of $^{39}K^+$ with energies in the range 400−1000 keV, concluded that the gas desorption from the wall surface is determined by the amount of energy given to electrons *inside* the material in a limited layer near the surface. They performed measurements with ion beams incident to the wall at different angles. For an ion trajectory at an 88° angle of incidence to the normal, where all electronic energy is released inside a very thin layer of ~ 150 Å touching the surface, they found a maximum release of 15000

gas molecules.

As a comparison, we can estimate the desorption coefficient for the YAGUAR experiment in the following way: with the gas desorbed density of $n_{H_2} \simeq 1.4 \times 10^{13}$ molecules/cm^3 obtained in the previous section, for our nn-chamber volume of 1130 cm^3 we have a total number $N_{H_2} \simeq 1.6 \times 10^{16}$. The YAGUAR gamma fluence per pulse is about the same as the neutron fluence, that is $F_\gamma \simeq 1 \times 10^{15}$ per cm^2 giving a total number of gammas $N_\gamma \simeq 0.8 \times 10^{18}$ incident on the nn-chamber surface of 800 cm^2. Therefore

$$\eta_\gamma = \frac{N_{H_2}}{N_\gamma} \simeq 0.02. \tag{3}$$

To understand whether or no such desorption yield sounds physics we will try to predict η_γ using recent findings of Molvik et al. [25] for $^{39}K^+$ ions. The study with potassium ions provides insights and data useful in understanding the desorption rate in the YAGUAR setup. Of interest is how deep below the metal surface the released electron energy influences the gas desorption process on the surfase.

Modeling Results

As a first step at predicting a desorption rate for the YAGUAR set up, we used a straightforward model to relate desorption yield to the energy deposited in a relevant layer of the aluminum surface. In our model particles enter the surface at some angle θ with respect to normal to the surface and travel in a straight line through the material with a total range, R. We treat each point along an ion trajectory as a possible electron source, assuming a uniform energy deposit along the ion track. To relate the maximum track depth in the aluminum, $z(\theta) = R\cos\theta$, to the number of desorbed molecules, we assume that each electron source point along the track produces a number of desorbed molecules at the surface that depends exponentially on the depth of that point, $\eta \sim \exp(-\ell/\lambda)$, where ℓ is the depth along the normal to the surface and λ represents a characteristic depth for the energy release below the surface which still significantly contributes to the desorption on the surface. In addition, as the angle θ is decreasing for a given energy particle, the total energy deposited along the track reaches deeper below the surface. Thus, the total energy available to cause desorption is spread through a thicker layer. Therefore, the strength of the local electron-source is weakend. Therefore, the desorption must also depend inversely on depth, $z(\theta)$. Integrating with respect to depth below the surface, we arrive at an expression for η as a function of incident angle,

$$\begin{aligned} \eta(\theta) &= \frac{\eta(90^\circ)}{z(\theta)} \int_0^{z(\theta)} \exp^{(-\ell/\lambda)}\, d\ell \\ &= \frac{\eta(90^\circ)\,\lambda}{z(\theta)} \left[1 - \exp^{(-z(\theta)/\lambda)}\right]. \end{aligned}$$

We can use this model to determine the characteristic depth, λ, from existing measurements which have known energy losses. This gives us a benchmark against which we can predict desorption rates for the YAGUAR setup.

Figure 3: Points are desorption rates determined by Molvic et al. [25] and Bieniosek et al. [26] for $^{39}K^+$ ions incident on stainless steel. Lines are calculations using the model described here for different values of λ: small dashed line ($\lambda = 400$ Å), solid line ($\lambda = 750$ Å), large dashed line ($\lambda = 1500$ Å).

Figure 4: The depth distribution of energy deposit per layer λ per incident gamma ray in a 2-mm slab of Al for 1-MeV gammas (boxes) and the YAGUAR gamma spectrum (diamonds). The energy deposition is simulated by GEANT4. The YAGUAR gamma spectrum used in simulation is from MCNP [28] modeling of the reactor core.

Molvic et al [25, 26] obtained desorption data for a 972 keV $^{39}K^+$ ion beam incident a stainless steel target as a function of incident angle. With these data on has $\eta(90^\circ) = 15,000$. Using these data and Eq. 4 we determine, as shown in Fig. 3, an optimal λ of about 750 $\mathring{A}$. At this depth $\eta(\lambda) = 9500$ molecules/ion. Furthermore, the range of $^{39}K^+$ in stainless steel is $R = 3914$ $\mathring{A}$ during which 82% of the energy loss is electronic (18% nuclear) [29]. Therefore, we find an approximate energy loss per particle by assuming all of the electronic energy loss (797 keV) is deposited in a depth of λ, giving 84 eV per the desorbed molecule and thus

$$\eta \sim \frac{\Delta E_\lambda}{84 \text{ eV/molecule}}, \tag{4}$$

where ΔE_λ is the energy in eV deposited in a depth λ.

For the nn experiment, we need to simulate the depth distribution of energy losses in the aluminum wall under gamma radiation. GEANT4 [27] was utilized for this purpose, with gammas incident on a pure 2-mm aluminum slab. In Fig. 4, a constant 1 MeV gamma source is compared to the more complex YAGUAR reactor gamma energy spectrum.

Assuming the λ for aluminum would be within a factor of two to that of stainless steel, we are interested in the energy deposited in the layer 750 to 1500 $\mathring{A}$ below the aluminum surface. Results show that $\Delta E_\lambda = 0.3{-}0.7$ eV are deposited in this region. Using Eq. 4 for this range of values to determine η_γ for aluminum gives a range η of $0.004 - 0.008$. These values are based on the $^{39}K^+$ data for baked stainless steel. It is well known that baked aluminum typically displays about a factor of five larger desorption than the stainless steel. Therefore, for the aluminum vacuum pipes in the current YAGUAR experiment we could expect desorption rates in the range

$$\eta_\gamma = 0.02 \text{ to } 0.04, \tag{5}$$

which should be compared to the value of $\eta_\gamma = 0.02$ estimated from the YAGUAR nn data.

Conclusion

Initial nn-measurements imply radiation-induced desorption of H_2 and possibly H_2O in the nn-collision cavity. Our model relating electronic energy deposit along the depth in

the target gives a value for the desorption yield η_γ close to the value assumed from the nn data. The next logical step is to minimize the desorption effect as much as technically possible. The DIa_{nn}A experiment currently has a signal to noise ratio of 1:40, which means to minimize this source of noise to the 10% level, we need to reduce the desorption effect by a factor of 400. Baking the aluminum (effectively removing the water content) can decrease desorption by a factor of 10. Irradiation by argon ions (at a relatively high flux of 10^{18} per cm^2) can decrease the effect by two orders of magnitude. New coatings are the most promising, with desorption yields reduced in 316 LN stainless steel coated in TiZrV and baked at 300°C by a factor of over 300. Further GEANT4 simulations of the realistic YAGUAR geometry will help in understanding the details of the energy deposit and may lead to designs to mitigate the influence of the low-energy electrons produced in the layers near the Al surface.

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